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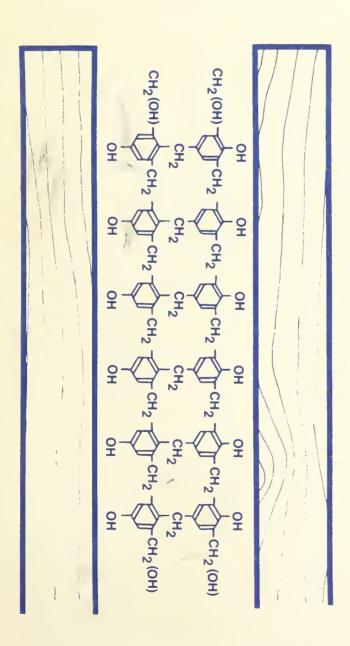
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PROCEEDINGS OF 1975 SYMPOSIUM....

# ADHESIVES FOR PRODUCTS FROM WOOD



Mr. . Mr. . La Calla

1976

FOREST SERVICE
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#### In Explanation....

The 1975 Symposium on "Adhesives for Products from Wood" was sponsored by the Forest Products Laboratory, Forest Service, U.S. Department of Agriculture. It was held September 24-26, 1975 at the Wisconsin Center Building on the campus of the University of Wisconsin.

Originally there was no intent to record the information from the meeting. But, due to repeated requests, this summary was put together. It does not include every paper; some of these presentations were made by specialists sharing research work in progress or material recorded in other publications. In some cases the papers will be available from the authors. However, their brief summaries generally indicate the tenor of their remarks.

Now that the various threads are pulled together, I hope it provides a background for future conferences-just as the records of the 1960 conference are valuable to us now.

U. S. DEPT. OF AGRICULTURE

JUN 1 7 1977

CATALOGING - PREP.

R. H. GIELESPIE, In charge of Improved Adhesive System Research Forest Products Laboratory and Symposium Chairman

# 1975 Symposium on ADHESIVES FOR PRODUCTS FROM WOOD

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1975 SYMPOSIUM "ADHESIVES FOR PRODUCTS FROM WOOD"

## The Chairman's Wrapup.

At the Symposium, a panel of R. F. Blomquist, A. A. Marra, T. E. Brassell, and R. E. Kreibich summarized their views on information presented and problems that must be solved. This led to an open exchange of comments about a variety of topics and viewpoints. Following is a condensation of these discussions:

Since the last Symposium 15 years ago, much progress has been made, but some of the same problems are still with us. The prospect of continued progress in wood adhesives is bright as evidenced by the attendance at the Symposium of many young people interested in adhesives, by the rapidity with which the chemical industry develops new materials, and by the new instruments and techniques for evaluation that are now available. Adhesive development, something of a black art in the past, is emerging as a technology with a firm scientific basis. Much of this advancement can be attributed to better analytical tools, including infrared spectroscopy, nuclear magnetic resonance, and differential scanning calorimetry. The application of the principles of fracture mechanics holds promise of providing a better insight into the relationship between the micro and macro structure of an adhesive and its performance in joints.

Little, if anything, was mentioned about nondestructive testing during the symposium, but not because the topic was considered unimportant. Future meetings should include information on this subject.

The Symposium was structured around current problems, particularly adhesives from renewable resources. Adhesives from tannins and lignins were discussed as well as bonding without adhesives, procedures still in their infancy. The reports presented on the future

of petrochemicals for adhesives were encouraging and indicated that the chemical industry had much to offer for wood adhesives regardless of the original source of the raw material. Continued research on resins from acetone might be beneficial since acetone price and availability look attractive for the future; a mole of acetone is produced with every mole of phenol, and phenol production is expected to remain high. Another raw material potentially attractive for adhesives is dicyclopentadiene, and research with this monomer might prove productive.

Several presentations dealt with predicting durability. Its importance and the need for additional research was emphasized during the discussion by such statements as: "If we had a new adhesive at 2 cents a pound that worked well in production and produced high-quality bonds with ease, no one would use it until its long-term durability were assured."

A plea was made to adhesive developers to place material on outdoor exposure at the earliest point feasible during the development of a new adhesive. Even the data from 1-2 years of exposure can be useful when the new adhesive is introduced into the marketplace.

More interest exists now than ever before in new and improved binder systems. There is more need for bonding in the wood industry, and the need will continue to grow as new products are developed and our timber resources are used more efficiently in either new or old products. The need is for more economical, more dependable, and more durable adhesives.

In the last 15 years we have recognized that adhesives are engineering materials. The mechanical properties of an adhesive (its modulus, creep, and strength characteristics) may govern the performance behavior of a bonded assembly. Adhesives based upon elastomeric materials are more readily deformed than the rigid conventional wood adhesives whose main function was to transfer stresses between adherends. The bonding of plywood to joists with construction adhesives is the first major use of these relatively low-modulus materials. Before this use can be expanded into other building applications, extensive investigation will be necessary into the mechanical properties of this class of adhesives.

Many roadblocks obstruct the development of new adhesive systems and new bonded products, and points of view differ about how such developments can take place. The users of adhesives keep pressing researchers and adhesive manufacturers to develop better and cheaper adhesives for

their product. They promise an established large volume as the incentive to invest in such developments.

But new products often exist only in some researcher's mind, and this gives rise to a multiplicity of problems. No one wants to develop a special adhesive, design and build special equipment, or attempt to make a market survey for a product that doesn't yet exist as a potentially salable item. Such a development would benefit by a team effort where adhesive formulators, equipment designers, and market analysts would provide special expertise to help unravel the complexities of new product development.

Another problem area for adhesive development is for existing products that require high-performance adhesives, but the total volume of use may be small. An example of such need is in fingerjointing for structural applications. One cost analysis had indicated that the adhesive for a fingerjoint involved only about 1 percent of the total cost of the joint. Most of the cost involved material handling, cutting, adhesive spreading, and radio-frequency curing. Fingerjointing might benefit from a more expensive or a more sophisticated adhesive if its use would reduce peripheral costs. But the volume requirements for such a specialty adhesive provide little incentive for investing development dollars.

Some representatives of adhesive manufacturers felt they needed much more information about adhesive needs than the user is able or willing to provide. The adhesive manufacturer wants to know about performance requirements, potential volume, and cost effectiveness--not just adhesive cost, but the total cost of bonding. He needs a value analysis upon which to base decisions about spending scarce development dollars. He wants to spend these dollars on work that has the greatest potential to pay off. The lack of suitable tenformation prevents adhesive manufacturers from tackling some of the more complex bonding problems that adhesive users propose.

One point of view was that adhesive developers were way ahead of engineering development in the industries using adhesives. Excellent adhesives are available with gel times as short as 12 seconds but the pieces to be bonded can't be handled fast enough with present materials-handling equipment. Production people must be willing to accept more sophisticated bonding systems and provide the plant lay-out and material-handling capabilities to take advantage of this new adhesive technology. These new techniques provide the opportunities to build houses as cars are built in Detroit and use adhesives to good advantage.

The fingerjointing problem stimulated discussion about possible solutions using somewhat exotic adhesive systems. The need is for a durable, gap-filling adhesive that would cure in seconds without radio-frequency energy. It would be preferable to avoid cutting fingerjoints and simply use butt joints, but this appeared beyond current technology.

It was suggested that certain epoxy systems will cure almost instantly with some polymercaptans. Certainly, a high percentage of ultimate strength can be achieved in seconds. Here metering and mixing is not the problem, but adhesive spreading and assembly of pieces is. This system, as with most others, cannot provide a few seconds press time and still accommodate several hours of assembly time.

This brought up the question about the possible use of hot melts for fingerjointing, but not the kind that will resoften when the product is used in hot areas. A potential candidate is a polyurethane that can be gap-filling and provide good adhesion. It might be applied as a hot melt but later react with moisture to cross link in a room temperature cure. Hot melts might be reinforced with chopped glass fibers to reduce creep or new ones developed that had higher glass transition temperatures. Current efforts among adhesive manufacturers are being devoted to the development of such bonding systems.

The question was raised about whether or not radiation-initiated polymerization might play a role in adhesives technology as it now does in coatings. It was proposed that radiation could take place with the pieces in the open position just after the adhesive had been applied. After radiation, the pieces would be assembled and clamped. This would overcome the major problem that radiation (from U.V. sources, cobalt-60, linear accelerators, etc.) penetrates wood only to a very limited extent. Use of external radiation sources is usually a more costly way of generating free radicals than by chemical means. Radiation polymerization can cure in a matter of seconds and has potential for specialty applications, but most attendees felt it unlikely to be used commercially in the immediate future.

Past research has demonstrated that it is possible to make products for structural uses starting with wood fibers, making waterproof liner-board and corrugating medium, and combining them with a waterproof adhesive. The problem is to develop an adhesive that will produce the instantaneous bonding in today's corrugating machines at the high speeds necessary for economy in production. The need was for an adhesive that would provide a quick-grab bond at room temperature but later cross link to a waterproof state. It was stated that a suitable adhesive

exists now, but someone must be willing to pay for the adhesive and to modify machines to handle it. Changing economic situations and a team approach to problem solving might generate a breakthrough on this problem.

In general, two areas of interest received the most attention in the comments by the attendees: 1. The need for continued research on durability prediction, and 2. The need for better communication and information exchange between all parties concerned with the development and use of adhesive technology.

The 1975 Symposium on Adhesives for Products from Wood was viewed by the majority of attendees as an effective medium for improved communications. In fact, the Symposium committee was urged to plan future meetings on a much more frequent schedule and not wait 15 years before the next one.

R. H. Gillespie

#### Session A, Wednesday, Sept. 24

Moderator: R. H. Gillespie, FPL, Symposium Chairman

Welcome: R. L. Youngs, Director, FPL

Keynote address--Review of 1960 Symposium, Its Predictions and Recommendations: R. F. Blomquist, Forestry Sciences Lab

#### Reports From Wood Bonding Industries

Adhesives in the Plywood Industry -- Alan Lambuth, Boise Cascade

Adhesives for Particleboard -- Charles H. Hickson, Borden Chemical Co.

Adhesives for Glued-Laminated Timber--T. E. Brassell, AITC

Adhesives for Housing, A Workshop Report -- R. H. Gillespie, FPL

Adhesives in Furniture Joints -- R. F. Snider, Franklin Glue Co.

#### Session B, Wednesday, Sept. 24

Moderator: W. F. Lehmann, FPL

#### Reports from Wood Bonding Industries

Adhesives for Composites -- Alan A. Marra, Univ. Massachusetts

Adhesives for Overlays and Patching--W. W. McCann, Simpson Res. Center

#### Adhesives from Nonpetroleum Resources

Bonding Characteristics of Acidified and Spray-Dried Spent Sulfite Liquor--K. C. Shen, Eastern FPL, Ottawa

Bark Extracts as Bonding Agent for Particleboard--Arthur B. Anderson, retired, FPL-Univ. California

Wattle Tannin Adhesives for Wood Bonding--H. M. Saayman, Leather Industries
Research Institute

Bonding Wood Without Adhesives -- William Johns, FPL-Univ. California

Amylaceous Phenol-Resin Extenders for High-Density Particleboard--R. K. Krueger, Krause Milling Co.

#### Session C, Thursday, Sept. 25

Moderator: B. H. River, FPL

#### Durability of Adhesives

Preliminary Correlation Between Weathering Tests and Automatic Boil Machine Results--Dick Caster, Weyerhaeuser Co.

Precision of Rate-Process Method for Predicting Life Expectancy--M. A. Millett, FPL Relationship Between Rate-Process Method and Weathering Tests--B. H. River, FPL Outdoor Weathering of Plywood and Composites--R. C. Raymond, American Plywood Assoc Softening Temperature Relations to Adhesive Durability--S. Chow, Western FPL, Vancouver

Reappraisal of the Durability of Melamine Resin Adhesives--R. E. Kreibich, Weyerhaeuser Co.

#### Session D, Thursday, Sept. 25

Moderator: J. A. Koutsky, Chemical Engineering, UW-Madison

#### New Developments in Adhesives and Bonded Products

Influence of Extractives in Dipterocarp Veneers on the Cure of Phenolic Adhesives--J. D. Wellons, Oregon State Univ.

Modified Melamine Resins for Producing Particleboard--W. Clad, BASF, Ludwigshafen

Developments in Contact Cements--B. R. Garrett, Roberts Consolidated Industries

Developments in Hot Melts, Emulsion, and Solvent Systems--W. C. Kania, National Starch and Chemical Corporation

Developments in Polyurethane-Type Adhesives--E. R. Harrell, 3-M Company

Adhesive Developments in New Zealand--D. V. Madle, ICI New Zealand Limited

Use of Adhesives in Construction, What's Next?--J. D. Rose, American Plywood Assoc

#### Session E, Friday, Sept. 26

Moderator: J. W. Koning, Jr., FPL

#### Forecast of the Future

Fundamental Research on Adhesives--J. A. Koutsky, UW-Madison

Future Expectations for Bonded Wood Products--T. H. Ellis, FPL

The Future of the Adhesives Industry--H. E. Roth, E. I. DuPont de Nemours and Co., Inc.

Future of Petrochemicals for Adhesives -- J. B. Toogood, Union Carbide Corporation

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#### OBJECTIVES OF MEETING

Like the initial symposium in 1960, this symposium is intended primarily to improve the lines of communications between present and potential users of adhesives in the wood products industry, the present suppliers of a wide variety of adhesives, the chemical industry which provides the basic components of adhesives, and the manufacturers of equipment used in the production of bonded wood products.

The principal focus is on what we now have available in adhesives, what our current adhesive-bonded products are and how they are made, what new products are anticipated, what new adhesive systems are required that are not now available, what the possibilities are to produce such adhesives and processes, and what production equipment is needed to use such adhesives more effectively in future production of wood products.

This symposium is planned for a maximum of discussion and audience participation. Formal presentations are limited to outlining current status of needs, available adhesives, to suggest some needed development of new adhesive bonding systems and processes, and some projections for the future. These presentations are intended primarily to stimulate discussions and contributions from the audience. The success of the symposium will depend largely upon the effectiveness of these audience contributions. Speakers are not expected to have all the answers. They are mainly to indicate the current state of the arts and to ask the basic questions. It is hoped that some of the important answers will be provided by those in the audience.

Many questions will undoubtedly not be answered this week. One important aspect of the symposium is therefore to summarize the unsolved problems and any potential solutions suggested. A review committee has been designated to summarize the presentations and discussions at the close of the symposium, and particularly to summarize the unanswered problems that need future research and development, and suggest priorities for these efforts. This material will be presented at the final session, as was done for the 1960 symposium.

Your active participation in the discussions is needed. There has to be two-way communication between speakers and the audience, and particularly between individuals in the audience.

#### Keynote Speaker -- Dr. RICHARD F. BLOMQUIST

Dick is in Charge of Housing Research at the Forestry Sciences Laboratory, Athens, Ga., a research unit of the Southeastern Forest Experiment Station, Forest Service, USDA, Asheville, N.C. He is also Professor of Forest Resources, and Adjunct Professor in the Graduate School, University of Georgia. From 1942 to 1966, he was Supervisory Chemist in Charge of Glues and Glued Products Research at the U.S. Forest Products Laboratory, and also served as Lecturer at the University of Wisconsin. He received M.S. and Ph. D. degrees in organic chemistry from the University of Iowa in 1936 and 1937, after receiving a B.A. degree from Coe College in 1934.

Dr. Blomquist is a past president of the Forest Products Research Society. He is also a member of the American Chemical Society, Society of Wood Science and Technology, a Fellow of the American Institute of Chemists, and a Fellow of the International Academy of Wood Science. He has authored numerous technical papers on adhesives for wood and for metals, on glued products, and on use of wood in housing.

In 1960 he was chairman of a similar symposium on adhesives at the Forest Products Laboratory. It is logical to have him review some of the highlights.

# REVIEW OF THE 1960 SYMPOSIUM, ITS PREDICTIONS AND RECOMMENDATIONS

The 1960 Symposium on Adhesives for the Wood Industry was primarily intended to provide better communication between wood industries that would need adhesives in the future, and the adhesive and chemical suppliers with a potential range of products to satisfy these needs. To a considerable degree this seems to have been successful.

In 1960 it was evident that the variety and volume of adhesives used in the wood industry were extensive, and their use has expanded greatly. Today the need for more new composite products of wood and wood-base materials is tremendous and adhesive bonding is the key to their successful development and production. The softwood plywood industry has increased threefold since 1960, including the establishment of the extensive southern pine plywood industry. Particleboard manufacture has increased greatly. Now we are on the verge of producing exterior-type structural panels and veneer-faced particleboard panels to supplement all-veneer plywood and to make better use of our residues. Successful use of composite wall studs in light-frame house construction has been demonstrated, and wider dimension lumber of composites is being investigated. Adhesives have played a great part in the wood industry in recent years, and they will continue to be even more important in the next 10 years.

The earlier symposium produced a series of 11 specific recommendations for future action. These are included in the following section. We have considered nearly all of them, and generally have made significant progress on many. But progress has been incomplete. Objectives have not been fully met. There is still a considerable way to go on each item.

Although many new adhesive systems have appeared for other bonding applications since 1960, few new systems have been introduced in the wood industry. We are still using the same types of adhesives for plywood, laminated timbers, furniture, and as particleboard binders as before. There has been a trend to the more durable adhesives for severe service, and many technical improvements have been made to meet individual plant production needs. The mastic construction adhesives have been extensively adopted by builders, particularly for the field-glued floor system. The wood industry still needs adhesive systems that are faster curing, simpler to use, more durable and economical than those currently available. The energy situation demands that we look to sources other than oil- and gas-based chemicals for adhesive components. Identification of specific properties needed in adhesives for new processes and products is vital in developing the necessary new systems.

The adhesive industry has grown extensively since 1960, and public confidence in bonded constructions has increased significantly. We need to adapt the sophisticated adhesive technology developed for applications outside the wood industry to our own needs now. To do this, we need to know each other's problems and potentials. This will be the theme of the present symposium.

#### RECOMMENDATIONS FROM THE 1960 SYMPOSIUM ON ADHESIVES FOR THE WOOD INDUSTRY:

Recognizing that better gluing processes are vital to continued improvement in the utilization of wood, and that a single all-purpose wood glue is still probably far distant, it is recommended:

- 1. That the wood industry make a continued and concerted effort to define more clearly the requirements for glues in specific gluedwood products.
- 2. That adhesive manufacturers continue their efforts to familiarize themselves with the changing requirements of the wood industry. They are urged to evaluate new raw materials, such as new polymers and catalyst systems, and new technology developed from other adhesive applications in meeting requirements for applications to wood.
- 3. That manufacturers of processing equipment be encouraged to take a more active interest in the development of improved equipment for all phases of the gluing process.

- 4. That there should be closer cooperation between wood-product manufacturer, glue manufacturer, and equipment manufacturer in contribution to research and development programs needed to produce new and improved wood products.
- 5. That continued attention be directed toward the development of new and improved glued products of wood and of combinations of wood with other materials in the interest of the economic utilization of our forest resource.
- 6. That greater attention be directed toward research on the requirements and performance of binders in particleboard and other molded wood products. A better understanding is needed of relationships between processing variables and board properties.
- 7. That fundamental research be undertaken to determine more precisely the nature of the interface between glue and wood surfaces of various species and of modified woods. More research is also needed to understand the interaction of glue and wood as related to the chemical structure of glue components and the nature of wood substance. The rheological properties of the glues themselves and of the final glueline should be studied further for better understanding of joint formation and behavior. Fundamental work should be undertaken to understand the nature of stresses in glued joints under different conditions of joint geometry and under various types of loading.
- 8. That more research be undertaken to understand the fundamental nature of the deterioration of glued joints and of glued-wood products in service. Every effort should be made to develop reliable short-term test procedures to ascertain the probable permanence of glues in joints on a performance basis, and to insure that these tests correlate adequately with actual service experience.
- 9. That research be continued to develop improved quality control tests for glued-wood products.
- 10. That greater effort be made to develop improved specifications for glues and glued-wood products so that quality may be properly safeguarded without excessive and unrealistic performance requirements.
- 11. That continued attention be given to the exchange of current views and research findings in the field of new adhesives and glued-wood products. That conferences, such as this symposium, be held at regular intervals.

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#### WOOD BONDING INDUSTRIES

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#### ADHESIVES IN THE PLYWOOD INDUSTRY

There are now more than 400 plants in North America manufacturing various kinds of plywood panels. In 1974 these plants produced about 21 billion feet of plywood (surface measure or 3/8-inch basis), and consumed about two billion pounds of mixed glue. Practically all this plywood was bonded with synthetic resin adhesives, phenolic for softwood and urea or melamine for hardwood.

As well as these glues serve the ever-expanding plywood industry, their use has created a condition of almost total dependence on the availability of three basic chemicals—phenol, urea, and methanol. Although we now realize the vulnerability of this position, completely acceptable alternatives for these critical chemicals and resins have not yet been found. Mainly as a result of the 1974 petrochemical shortage, protein adhesives for plywood have made a modest comeback.

Due to the steadily rising cost of labor in recent years, much effort has been directed toward automation at every step in the assembly and handling of plywood. Glue application methods have diversified from the hand-fed hard roll spreaders of 1960 to the sprays, curtain coaters, extruders, and in-line soft roll applicators of today. Prepressing is now almost universal. As a result of these changes, the plywood glues of recent years have had to feature exceptional versatility to fit the requirements of new manufacturing methods. Substantial improvements in assembly time tolerance have been made with minimum impact on prepressing or cure time.

Assisted by the APA's broadened adhesive composition limits and a generally upward trend in both spreads and solids, today's resin glues are able to satisfactorily bond the tough veneer from small-diameter logs and most of the woods being imported to extend our domestic supply.

# Adhesives in the Plywood Industry

#### Alan L. Lambuth

In the summary you were given there are some fairly impressive statistics relating to the plywood industry of North America. It now includes about 425 plywood mills supported by an additional 200 veneer plants. Slightly over half these plants make softwood plywood while the rest manufacture hardwood panels. This fairly even division in numbers of plants is somewhat misleading in terms of capacity, since the softwood mills produce 18 billion square feet 3/8-inch basis, to about 3 billion square feet surface measure from the hardwood mills. Collectively, these plywood mills consumed about two billion pounds of mixed glue in 1974. These figures represent best estimates from several association, industry and government sources.

Practically all these two billion pounds of plywood adhesives were based on thermosetting synthetic resins made from phenol or urea and methanol. Phenol-formaldehyde and to a lesser extent melamine-formaldehyde resins are used to produce all exterior grade plywood. Urea-formaldehyde resins are used to make virtually all interior hardwood panels, while phenolic resins at various levels of extension produce all but a small portion of the interior softwood panels for structural purposes.

These synthetic resins actually serve the adhesive needs of the plywood industry very well. In fact, on a cost-performance basis, they have no present equal. This has resulted in a condition of almost total industry dependence on the three basic chemicals, phenol, urea and methanol. While there are several natural sources for each of these compounds, they are most easily and cheaply produced in the volumes required from petrochemical intermediates. Thus, the plywood industry is at present almost totally dependent on the world's crude oil resources, with all the hazards and uncertainties this represents.

Although projections of price and availability for our three basic chemicals are reasonably favorable over the next several years, barring major changes in world economics and markets, we do face long-term supply problems relating to the adhesives we use now. Research into alternate polymers and adhesive systems for bonding wood is under way in several parts of the world. Considering the magnitude of our ultimate problem, these research efforts deserve our wholehearted support now.

Meanwhile I will discuss phenolic and urea glues as though they were here to stay.

<sup>\*</sup> A general interest paper presented at the U.S. Forest Products Laboratory's Adhesive Symposium in Madison, Wisconsin, on September 24, 1975.

#### Glue Line Performance Levels

For structural softwood plywood, three separate levels of glue line durability are presently specified in U.S. Product Standard PS1-74. These are interior, intermediate and exterior. All three levels must have strong, permanent and creep-resistant dry bonds to insure adequate structural performance under fixed and variable loads. In addition, all performance levels must not delaminate while being destroyed by fire. Delamination under intense heat contributes fuel and causes premature structural collapse. Thus, the differences in durability really only apply to weather or water resistance. For instance:

Interior plywood, intended for covered applications, has only moderate water resistance to withstand cycling humidity and occasional wetting.

Intermediate plywood is basically a "super interior" grade which is intended to withstand considerable job-site weathering before going into covered service. It will also take repeated wetting after installation.

Exterior plywood is bonded to give long-term service under continuous outside exposure, immersion or cyclic wetting and drying.

Marine and siding panels are simply special exterior grades with higher quality cores supporting the exposed face veneers.

Hardwood plywood is also produced in several durability grades reflecting end use requirements:

Type I is generally equivalent to Exterior softwood plywood, while Technical grade approximates Marine Exterior, having higher grade inner plies. Both types of hardwood plywood must meet appropriate levels of dry shear strength and boil shear wood failure.

Type II is general purpose moisture resistant interior plywood with no particular limitation on service life.

Type III is moisture-resistant interior plywood for limited life applications such as crating.

Parquet flooring made from hardwood veneer is now covered by Type II rather than Type I durability requirements due to a fairly recent limitation on below-grade installations.

In addition to these performance levels which are controlled and approved by the respective trade associations, individual plywood plants can and do market a certain amount of plywood as "mill certified" for the intended use. These are generally lower durability product applications.

#### Plywood Manufacture

Two resource-related events of the nineteen sixties had major impacts on the methods of plywood manufacture and even the glues that were used. One was the creation of a southern pine plywood industry. The other was a general swing to sustained yield softwood log supply. Both were stimulated by (and dependent on) the development of high speed small log handling equipment at the green end of the plant. Once it became possible to charge and peel several small logs a minute, the flow of veneer through the rest of the plant equalled the rate from bigger logs and production capacity was sustained at economic levels.

The success of these green end developments, plus the steadily rising cost of labor led to a reevaluation of manufacturing methods through the rest of the plywood process. Layup, pressing and panel finishing proved excellent candidates for at least semi-automation. From this momentum for process change has come a variety of new plywood assembly and handling methods, and even new panel constructions. For instance, four and six ply layups were quite literally unheard of in 1960.

#### Application Methods

Automated plywood assembly methods frequently require new glue application techniques. Thus, the hard roll spreaders of earlier days are being steadily replaced by curtain coaters, sprays, extruders and soft-roll in-line applicators. In the 54 plants comprising the Southern pine plywood industry, glue application is now about equally divided among curtain coaters, spray lines and spreaders, mostly soft roll. On the West coast, hand-fed spreaders still account for about two-thirds of all plywood production, but the use of these other application methods is increasing. Glue application in Canadian mills is still mainly by spreader.

One philosophical comment is in order. If progress in plywood production is the goal, automation is not necessarily all good and hand operation all bad. There are bushelling spreader crews still operating in our industry that can steadily whip any automated line ever invented on a strict cost-performance basis. These are closely knit crews of dedicated and highly paid men with piece work as an incentive. Where crews of this type are available in the local work force they are worth serious consideration as "semi-automatic" production units.

#### Layup and Pressing Conditions

#### Phenolic Glues

In spite of the new layup procedures and glue application methods which have been introduced during the last decade, only a few of the working limits under which plywood is assembled and pressed have changed significantly. For instance, West coast glue spreads have increased a few pounds from mid-sixties to low seventies per MDGL, primarily to offset the increasing roughness and toughness of veneer from today's smaller diameter logs. Assuming a uniform level of glue solids, the spreads tend to remain fairly constant regardless of the method of application, whether, spray, curtain, roll or extruder. Southern pine glue spreads are 10 to 15 pounds higher, of course, due to the greater absorptive properties of the species, but again tend to be constant among the methods of glue application.

Prepress cycles have been somewhat shortened since the introduction of this technique in the early sixties, but not greatly. Two minutes' top panel stand time plus two minutes under 150 psi pressure will generally produce a good bond on any softwood, using a glue mix intended for the purpose.

Selecting 5-ply 13/16-inch panels as a basis for comparison, 5 1/4 minutes at 300°F would be considered a fast press time, 6 1/4 minutes average and 7 1/4 minutes slow by today's as well as earlier standards. There has been a shift in emphasis from "fast" to "average" times in order to accommodate wider assembly time limits. The capability to provide fast press times is equally available today, however.

The big change has been in maximum assembly time limits. APA-approved assembly times have increased from an average of 20 minutes in 1960 to 45, 60, even 120 minutes now. This change in adhesive capability has greatly simplified the introduction of automated layup procedures and hot press production scheduling. Minimum assembly time limits have remained unchanged at one to three minutes.

The range of hot press platen pressures is still dependent on wood specie; 100 to 125 psi for crushable species like cedar up to 200 or 220 psi for dense woods, or species with very wide summer wood rings like southern pine. 175 psi is still the most frequently used pressure for mixed western softwoods.

#### Urea Glues

The layup procedures, conditions and limits for assembling hardwood plywood with urea resin glues have changed relatively little over the last decade. In particular, the precision required in the layup of decorative hardwood panels has fairly well precluded the introduction of faster but less accurate assembly methods. Glue application remains almost exclusively by spreader.

Urea glue spreads have been fairly constant for many years at about 60 to 75 pounds per MDGL on softwood cores and 75 to 90 pounds on hardwood cores.

Although urea glues can be acid-catalyzed to cure faster than phenolic glues, their press times are generally similar. Instead, pressing temperatures are lowered to minimize blistering. Six minutes at .260°F would be average and 5 1/4 minutes fast for a 13/16-inch five-ply hardwood panel glued with UF adhesive. If a trend is discernable in hot pressing, it is toward slightly higher press temperatures in recent years; for instance, 275°F. instead of 240 or 260°. Temperatures of 280°F and above are occasionally used for special purposes.

Because of their greater inherent flow and water-holding capabilities, urea glues require much longer prepress cycles than phenolics. Twenty minutes' top panel stand time plus ten or twenty minutes under pressure is not uncommon. The compensation for these long preliminary cycles is the much greater assembly time tolerance which urea plywood adhesives exhibit. Total assembly periods of one to several hours after prepressing are feasible and generally used. This permits a considerable degree of latitude in scheduling loads of plywood through the press, including layup for one shift and hot pressing for two.

At the present time, there is no short-cycle cold press urea glue system for plywood comparable to the NC clamp process for protein cold press glues. A system of this type is available for urea door glues but not plywood. Thus, hardwood panels made with cold press urea adhesives must still be clamped with turnbuckles and irons for at least several hours.

#### Glue Formulations

Although straight phenolic and urea resins have established themselves as outstanding industrial adhesives for bonding wood fiber, flake and chip into board products, their use in plywood has not proved too successful. The principal reason, in my opinion, is that operating conditions governing the stock moisture content, applied resin solids, dwell time and hot pressing of board products can be controlled within fairly narrow limits, to which resin properties can be accurately adjusted. By comparison, the operating conditions in plywood manufacture are necessarily broad, even

contradictory, from the standpoint of resin performance properties. The answer for plywood has been to formulate the neat resins into mixed adhesives using ingredients which supplement and extend the performance of the resins to cover these broader requirements.

Next to resin itself, the extender used in a glue mix tends to establish its performance limits as an adhesive. Many years' experience has shown that a combination of extenders consisting of finely ground particulate material teamed with a glutinous dispersed material gave the best balance of properties over the normal range of plywood layup times. In theory, at least, the fine particles prevented overpenetration and joint starvation when the press is closed on the still-fluid glue lines of a short assembly time panel, while the glutinous material held water at the glue line for long assembly time transfer and flow, resisting the strong absorptive force of dry veneer. Since there was a fixed and fairly low limit on the proportion of glutinous extender permitted, the favored particulate extenders were those which would swell and disperse and contribute some additional water-holding properties to the glue also. The dispersion was generally accomplished with a strong alkali, usually sodium hydroxide.

This theory still characterizes current glue formulating practice. A higher level of glutinous extender has now been thoroughly tested and approved, relieving the particulate extender from the necessity of contributing water-holding properties, but the ground rules otherwise remain very much the same. Glufil plus wheat flour, Furafil plus sorghum flour, or bark flour plus potato starch would be typical and functional combinations.

Alkali is normally added to disperse the starchy material and swell the particles. For certain specialty adhesives, particularly those related to hardwood gluing, the caustic is often omitted, leaving only resin alkalinity to carry out the dispersion.

The rules for urea glue formulating are somewhat different in that much higher proportions of starchy extender are normally used. In fact, straight wheat flour extensions are quite common. The reason this can be done is that starchy materials like wheat flour are not fully dispersed in neutral or slightly acid solution, leaving a substantial portion of the mix as solid particles. Acid catalysts are then added to give hot or cold press cures.

#### RTU Glues

A special case in glue formulating are the ready-to-use plywood glues, mostly phenolics but also potentially urea adhesives. They are designed to be stable in viscosity, consistency and performance properties over a reasonable storage life. RTU glues are formulated by the adhesive supplier, generally along the lines outlined above, and are delivered to a plywood plant ready to store and use. There are now at least three plywood plants operating without glue lofts or mixing equipment, only storage tanks. Holly Hill is the first new plant actually designed and built in this manner.

RTU glues were originally developed as high solids adhesives which were approved for use at much lower spreads than normal and applied with special fine-grooved rolls. The idea was to cut down on water in the glue film in order to leave a higher tolerance for moisture in the veneer. The RTU concept has since been broadened to include the formulation of stable, storable glues down to about 27 percent resin solids. They are handled and applied conventionally. Because of their lower solids, shipping distance and cost become serious considerations.

#### Mixing Equipment

Glue handling and mixing techniques have remained much the same over the last decade or so with a couple of evolutionary exceptions:

- 1. There has been a trend toward fewer and larger mixes of glue, with as many ingredients as possible supplied in bulk and controlled by liquid flowmeter. This has permitted oneshift glue mixing for three-shift operation. Accordingly, both mixing and storage equipment have been scaled up in size, so that 5000 to 10,000 pound-capacity mixers and 50,000 pound storage tanks are no longer uncommon. The stability of today's plywood glues have made this development possible.
- 2. High-shear impeller mixers are steadily replacing the slower counter-rotating paddle mixers which have been widely used for so many years. These high shear mixers are fast, simple and efficient but their introduction has again been contingent on a glue mixing change. The heavy, dough-like initial dispersion step of earlier formulations has largely been replaced by a more fluid initial wetting and mixing stage. While the high-shear mixers could not handle the older doughy consistency, they efficiently homogenize the newer fluid first mixes.

#### Physical Properties

Unlike phenol-formaldehyde resins for paper impregnating, Bakelite molding and fiberboard, plywood phenolics are highly alkaline resin solutions containing 40 to 45 percent non-volatile solids. The solvent is normally all water. These resins are substantially more advanced than industrial PF's, having a molecular weight range of one to several thousand. Their starting viscosity is generally 500 to 1500 centipoises at 70°F. It rises slowly on 70° storage and doubles in a month or less. Given adequate heat and surfaces which absorb moisture, these resins polymerize to a complete, insoluble cure without the addition of catalysts, crosslinkers or curing agents. A rule of thumb for phenolic glue line curing temperature is one minute above 220°F. Once cured, phenolic resin glue lines are permanent.

Urea resins for plywood generally contain 50 to 65 solids in neutral water solution. The lower solids, more highly advanced resins definitely prepress better, at the expense of some assembly time tolerance. Resin viscosity is usually in the range of 500 to 1000 centipoises at  $70^{\circ}F$ . Except at the lowest solids levels they are stable on storage for one to several months at  $70^{\circ}F$ .

Urea resins require acidification in order to cure. Depending on the strength of acid addition, they can be made to cure cold as well as hot. This feature gives urea resins their well known versatility as wood adhesives for many purposes. In particle board, the acidity of wood alone is often sufficient to cure the UF binder. In plywood, with far less wood surface exposed on the glue lines, an acid-acting material must always be added. Unlike the phenolics, fully cured urea resin bonds are subject to gradual or rapid destruction by heat plus moisture. This behavior has prevented their use in structural plywood.

Melamine-formaldehyde resin, the durable but sluggish "big brother" of urea resin, is normally supplied as a spraydried white powder. It is stable for a year at 70°F. and is reconstituted into liquid resin by simply adding water. It requires both acid and heat to cure; in fact, much stronger acid additions than are needed for urea resins. Once fully cured, melamine resins are considered exterior durable. They produce the only white or low-color cured glue line presently available for exterior and high-moisture exposures.

#### Extenders

With very few exceptions, urea and phenolic resins are formulated into plywood glues by the addition of appropriate extenders plus water. The purpose is generally twofold; namely, to improve the performance and reduce the cost of the straight resin. A variety of suitable low-cost extender materials are available throughout North America. They are classified as follows:

#### Cellulosic

The dominant supply position held by Quaker Chemical's Furafil-based extenders through the late fifties and sixties has now diminished to about 50 percent of the market for several reasons:

- 1. Uncertainties in raw material supply have recently caused several changes in composition and performance (rice and oat hulls are being heavily substituted for the original corn cob content, and their behavior in mixed glues is significantly different.)
- 2. The recent trend toward higher adhesive solids has deemphasized the need for the unique thixotropic and water-holding properties of Furafil.
- 3. The higher levels of starchy extender now permitted by the APA have provided an alternate means for attaining some of these essential glue handling properties.

Especially in conjunction with higher amylaceous extender levels, there are now available a number of satisfactory alternate cellulosic extenders of appropriately fine grind for plywood glues. These include:

- 1. Nut shell flours
  Walnut
  Pecan
  Coconut
- 2. Tree Bark flours
  Douglas fir
  Alder
- 3. Wood flours
  Pine
  Fir
  Particle board sander dust
  Powdered Alpha Cellulose

All these extenders have been approved and used successfully in the plywood industry; some, like nut shell and wood flours, for at least 30 years. Blends of these and other extenders with Furafil are quite common, usually for cost reduction.

There are also a couple of wet cellulosic extenders now being introduced to the plywood industry. These include the "original" corn cob Furafil from a new source at 30 to 40 percent moisture content and paper pulp sludge fines at 50 to 60 percent M.C. The pulp sludge fines are claimed to be "pre-hydrated" and ready for use in both acid and alkaline glues without further dispersion.

#### Inorganic

- 1. Attapulgite clays
- 2. Bentonites
- 3. China clays

As low-abrasion thixotropic extenders the Attapulgite clays are now being used quite extensively throughout the plywood industry, generally in combination with cellulosic extenders. They can also be used alone. To attain intermediate levels of glue viscosity and thixotropy, the higher water-holding Attapulgites are blended with calcined Attapulgites or lower-cost China clays.

Although equally high in water-holding capability, the Bentonite clays have not been used much in recent years because of their significant abrasion level and tendency to form lumpy glues. Chrysotile asbestos fines, another formerly promising inorganic extender, have virtually left scene due to a potential respiratory hazard, even when resawing cured glue lines. Calcium-containing extenders like marble dust interfere with the adhesive properties of both urea and phenolic resins and should not be used in these glue systems.

#### Amylaceous

Starchy extenders play such an important part in the performance of urea and phenolic plywood glues that it seems improper to call them "secondary extenders". The implication is they are added solely for cost reduction or bulk. The fact is they generally cost more than the cellulosics and contribute rheological properties the cellulosics cannot duplicate. Among the more important of these properties are:

- 1. Greater assembly time tolerance.
- 2. Better resistance to overpenetration.
- 3. Faster and stronger prepress bonds.
- 4. Tackier consistency (more wipe resistance).
- 5. Deeper adhesion on dense woods.

Wheat flour is by far the most frequently used starchy extender in both urea and phenolic plywood glues. In general, the harder wheats and lower grade flour fractions are used in highly alkaline phenolic glues because they resist hydrolysis well and give a longer adhesive working life. The soft wheat flour, being more easily soluble and dispersible, tend to be used in slightly acid urea resin glues and moderately alkaline phenolics. There are exceptions, of course.

Other amylaceous extenders which have gained approval and moderate to widespread use include:

- 1. Sorghum flour
- 2. Potato flour and starch
- 3. Tapioca starch
- 4. Milo starch
- 5. Prepared or "instant" starches

The root starches, potato, tapioca and even yam, are particularly tacky and water-holding when dispersed. They actually require alkali for dispersion and remain stable for considerably longer storage periods than wheat flour. They are also widely used as paper corrugating adhesives because of their superior "grab". Prepared starches are mainly used in the manufacture of adhesives and packaged hardeners.

#### Protein

The principal protein additives to phenolic and urea glues have been spraydried soluble animal blood, usually beef or hog, and soybean flour.

A decade and more ago, spraydried blood was added to phenolic plywood adhesives (within APA-specified limits) to reduce press times and improve blister resistance. There were several significant drawbacks to using blood. The resulting glues were difficult to mix without lumping, they had limited storage life and equally limited assembly time tolerance. If fast press times were needed badly enough, however, one used blood and lived with its limitations.

With the current emphasis on bulk glue storage and greater assembly time tolerance, blood additions are seldom used today. The longer flow requirements of Southern pine plywood proved especially difficult for bloodfortified phenolic glues. The press time advantage they formerly offered is now largely obtained with newer and faster phenolic resins.

Blood additions to urea resin glues contribute more to durability than speed, and for that purpose are in current use. The blood is generally an ingredient in a blended hardener-extender package.

Soybean flour enjoys little current use in either urea or phenolic plywood glues except as a minor constituent in certain industrial wheat flour products. Virtually all its outstanding cold press adhesive properties are lost in hot pressing with resins, so regular wheat flour proves equally effective and cheaper as a water-holding additive.

#### Lignin Derivatives and Bark Extracts

In times of raw material shortage like the recent petrochemical crisis, a variety of wood-derived "phenol substitutes" or functional fillers tend to appear as partial replacements for whole phenolic resin or its phenol content. With the exception of certain well-known and widely used natural phenolic substances such as wattle, mangrove or quebracho extracts, these alternate materials are passive to phenolic resin, or so low in chemical activity that they are passive for all practical purposes. Thus, they contribute little to adhesion and cure and should be classified more as extenders than resin solids. Like other extenders they can be tolerated up to a point. They may contribute to consistency and water-holding but not significantly to adhesion.

Several research groups around the world are making real progress toward the use of lignin from pulp mill waste as hot press binders for wood products. With one exception, these are acid-curing replacements for our alkaline phenolic resins and are not meant to be combined with them. The single exception is an alkaline lignin product from the Finnish Pulp and Paper Research Institute which reportedly can replace up to 40% of the resin in a glue mix without undue effect on wood failure or durability. Initial APA tests on a Finnish adhesive of this type showed a somewhat slow cure but exterior performance on Douglas fir veneer. Work is continuing in several promising areas of lignin adhesive development.

#### Dispersing Agents

No constituent in a phenolic plywood glue causes more controversy than the alkaline dispersing agent. Even though there is general agreement that industrial grade 50 percent sodium hydroxide solution is the cheapest and best to use, considerations of quantity, placement and need for other agents tend to start arguments anywhere.

Originally, the cellulosic and amylaceous extenders were digested in a doughy first mix with water, caustic, and possibly sodium carbonate. This <u>did</u> give the best possible dispersion and water-holding properties,

but required a heavy first mix and special mixer. Now, with the necessity for complete dispersion somewhat deemphasized, a fair proportion of the phenolic resin is added initially for fluidity.

The present range of alkali addition is about 10 to 20 percent solid caustic, based on the total weight of extender materials. 8 to 10 percent is considered "low" and 20 or 22 percent definitely "high" for purposes of extender dispersion.

The varying opinions about including five to ten percent sodium carbonate as an additional dispersant are typified by the following comments:

- 1. It limits the efficiency of caustic digestion.
- 2. It reduces blisters during hot pressing.
- 3. It interferes with prepressing.
- 4. It helps reduce press time.
- 5. It causes lumps in the glue.

On a cost-performance basis of experience, my own preference is to leave it out.

There is a modest opportunity to develop initial dispersing alkalinity in preparing urea plywood glues. Ammonia-based liquid catalysts are available which make the first water-wheat flour mix alkaline for a period of time before becoming acid on resin addition. Powdered hardeners with similar properties can also be obtained. The key is to add the urea resin last instead of first in order to let the wheat flour extender get a brief alkaline exposure. Prepress times are reduced and assembly time tolerance noticeably improved by this procedure.

#### Additives

Small additions of particular compounds will often improve a particular aspect of adhesive performance very significantly. Defoamers are a familiar example. No matter how well a glue may otherwise perform, its value is limited if it foams excessively.

Other useful but less dramatic additives include:

- 1. Wetting agents, to insure uniform glue coverage and intimate contact with sound wood in the veneer surface.
- 2. Cuetain-forming agents to maintain continuous falling glue films at lower spreads and head pressures. Blips, tears and shadows are thus minimized or eliminated. Curtain forming additives are quite frequently also wetting agents, so both benefits are obtained.

- 3. Prepressing agents to increase prepress bond strength and decrease cycle time. Complexing salts like borax are typical of these compounds. They usually shorten assembly time also. Some thickening agents have also been offered to improve prepressing.
- 4. Glue line identifiers to permanently identify plywood for sales or legal purposes. Among the materials which have been used successfully for this purpose are: flake graphite, powdered metals, salts of rare elements, mica flakes, fluorescent materials and colored chips of cured resin.
- 5. <u>Viscosity or tack-producing agents</u> to thicken glues or give them a stickier consistency.
- 6. Thixotropic agents to make glues thicken rapidly after application and more resistent to smearing and overpenetration.

These and other additives are frequently used by adhesive manufacturers to incorporate the desired properties in their various glues. However, adding them at the time of use for the purposes indicated permits a single resin or adhesive to be optimized for several different applications. Most additives are easily incorporated into a mixed glue.

## Protein Glues

Except for an isolated plant or two in the West, protein hot press plywood glues virtually disappeared from the North American marketplace during the sixties. There were several reasons:

- 1. Greater emphasis was being placed on plywood durability.
- 2. The very low cost of synthetic resins made it simpler to use only exterior phenolic glues for all products.
- 3. A growing worldwide demand for food and fertilizer significantly increased the price of soybean flour and dried blood and reduce their availability.
- 4. More plants were installing bigger hot presses, so the press time advantage of protein glues became less important.

More recently, the supply problems which plagued urea and phenolic resins during the oil crisis have stimulated a moderate interest in returning to protein plywood glues. Also, the cost of protein raw materials have come down as petrochemicals have gone up, so protein glues offer a fifty percent cost saving over exterior phenolic glues even today. According to APA figures, hot and cold press protein glues now represent about four percent of the total of softwood plywood adhesives consumed. This includes two mills using soybean cold press glue.

Today's protein hot press adhesives are mostly based on spraydried blood with lesser quantities of soybean flour and wood flour. Lime, caustic and sodium silicate are the dispersing agents. For a more complete discussion of protein glue formulation and properties, please refer to Irving Skeist's "Handbook of Adhesives", Reinhold Publishing Company, 1962.

### Environmental Effects

The principal effect of plywood glues on the environment is through contamination of the wash water from glue loft and layup area. This waste water usually contains unacceptable levels of:

Phenol and phenol-acting solids
Formaldehyde
Alkali
Fermentable proteins, starches and sugars
Suspended solids

With respect to life forms in streams or other bodies of water, this washdown effluent is both poisonous and high in competing biological oxygen demand.

Adhesive and equipment washdown water is now mostly collected, stored and consumed in subsequent batches of glue. In either settled and filtered form, or agitated and suspended form, this recycled wash water requires only minor glue formulation adjustments and otherwise causes no changes in adhesive performance. There is a one-half to one percent recovery of resin solids that were formerly wasted.

This recycling program has caused some restriction in the use of readyto-use glues, since they normally require no in-plant water addition. In some cases, RTU glue composition has been adjusted to permit the addition of suitable amounts of wash water.

Vapor emissions from the hot press during plywood adhesive cure are not yet considered a significant source of atmospheric pollutants. Because of their high level of advancement, phenolic adhesives give off almost no organic vapors. Even the formaldehyde evolved from urea adhesives during cure is relatively non-polluting due to the high concentration of photo-chemical formaldehyde already present in the atmosphere, having been converted from other hydrocarbons in, say, car and diesel truck exhaust.

Plywood plant pollution control and energy-saving measures like reburning dryer emissions and direct-firing veneer dryers with sander dust have caused no significant changes in adhesive requirements for bonding dried veneer into plywood.

# Industry needs

From all of this, what does the plywood industry need in the way of new adhesive technology and what is lacking that presently impedes its progress? Practically speaking, not too much, in my opinion.

Considering the fact that our dependence on thermoset adhesives will do nothing but grow as we are called on to create ever-increasing quantities of building materials from ever-diminishing quantities and quality of forest resource, our number one need has to be the assurance we will continue to get adequate amounts of phenolic and urea adhesives, or similar-performing alternates, at the same or lower price. Having received this assurance, we might request a few other technical niceties in the name of progress. These would include:

- 1. Gap-filling thermoset glues which would maintain strong, all-weather bonds while eliminating the necessity for uniform, closely fitting surfaces.
- 2. Cold press phenolic adhesives which would give the durability and cost of current PF glues at urea or resorcinol pressing conditions.
- 3. More adhesive thermoset glues which would bond permanently to a wider variety of imperfect wood surfaces, such as overdried, overaged, extractive-coated and possibly fungus-modified.
- 4. Longer flow, yet faster-curing phenolic hot press adhesives, possibly catalyzed. Ideally, these glues would reliquefy from a dried film in the absence of water before proceeding to a rapid lot cure.
- 5. Low-color thermoset adhesives to produce unobtrusive exterior glue lines at half or less the cost of melamine adhesives. Ideally, these would be both hot and cold-curing.

I would make one comment in closing. In the interests of brevity and overview I have omitted mentioning a number of special cases and exceptions relating to plywood glues and procedures. They exist to be sure and are properly the subject of a more detailed discussion of industry practices.

Thanks very much for your patience and attention.

#### CHARLES H. HICKSON

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#### PARTICLEBOARD ADHESIVES - PAST, PRESENT AND FUTURE

A brief history is presented of the beginnings of the particleboard industry and the adhesives used: Its birth and infancy in Europe and its growth in the United States and Canada. In a very short span of time this industry, in the United States, has grown from an infant into a giant.

A view is given of today's adhesive market and the many changes that have taken place in the manufacture of particleboard adhesives. A trend is shown toward the production of tailor-made resins because of the variety of particleboard processes and the many end uses of this product. The need for highly specialized resins imposes new parameters on the adhesive manufacturer.

Now is the time for a look into the future of this industry and some of the possible problems we could face in the coming years.

# PARTICLEBOARD ADHESIVES: PAST, PRESENT, AND FUTURE

Particleboard was first produced industrially in Bremen, Germany, in 1941. This production was short lived as World War II severely curtailed all work on this product due to the shortage of both wood and resins which were consumed in the war effort. After the war, particleboard was again vigorously pursued in West Germany.

The initial research and development of particleboard in Germany was directed toward utilization of woodworking wastes due to the extreme shortage of lumber and all wood in general. This made these woodwastes a valuable raw material, certainly much too valuable to burn for heat value.

During this same span of time, the English, Swiss, and other European countries were also pursuing the manufacture of this type product. America and Canada were also interested in this application, and in the late 1940's, the first industrial particleboard produced in the United States was manufactured in the state of Virginia. There were several pioneers in this country - a few of them were Swain Industries, U. S. Plywood Corp., and Miller Hofft Co. These were the first to note the economic potential of particleboard in the U. S.

The first boards, produced in Germany, were made of woodwaste and hogged fiber with anything but a controlled fiber size. This, of course, yielded physical properties that were not consistent and were far from satisfactory. German development of the wood "flake" in the late 1940's, that is the control of the geometric configuration of the chip by selecting the proper grinding process, allowed the production of a higher quality board than previously produced and led to the development of the type of board we see in the market place today.

In the United States, control of flake geometry, need for this particular kind of product, and the use of an economical adhesive were the three major building blocks that allowed the Particleboard Industry to grow and flourish at such a fantastic rate. Adhesive development centered around two major type resins — urea formaldehyde and phenol formaldehyde.

UF resin binders for this purpose were introduced by I. G. Farbinindustries around the same time that the early particleboard development work in Germany was getting underway. This resin is a condensation product of urea and formaldehyde. For its use as a wood binder, it is stopped at a stage in the condensation where the product is still water soluble and has the properties necessary to produce a good board and meet other physical and chemical requirements of the user. The reaction goes through the formation of mono and dimethylolurea followed by condensation. This can be considered a precursor in the formation of the final crosslinked or cured urea formaldehyde resin. The exact mechanism is still not known with complete certainty. During the condensation, water is formed, making an exact cofficulation of chemical solids impossible.

Phenol-formaldehyde resins also go back a long way. It was one of the first synthetic plastics manufactured. Probably Baekland did more than any other person to make them a commercial realization. Phenol and formaldehyde also go through an addition reaction to form methylol compounds. The methylolphenols further condense, splitting out water to form methylene linkages. These phenolic nuclei condense to give a crosslinked network. The condensation of these resins, as with the ureas, are also stopped short of completion so that the resin is still water soluble which, when applied to the chips and cured will produce a saleable product. Phenolic resins are more durable than the UF's and exhibit a resistance to breakdown in both hot and cold water. In addition, they are resistant to acid, alkali, and common organic solvents.

On the other hand, they do have several drawbacks in comparison with UF's. They do not exhibit the same degree of tack, are slower curing, have less liquid storage stability, and impart a dark color to the board. In cost, they are more expensive than the urea formaldehyde polymers.

Between 1946 and 1950, many different types of glues and adhesives were tried as binders for particleboard in the United States and Canada. Many of these glues worked but none showed the versatility nor the economic advantages of the UF resins. Around 1950, urea fermaldehyde as a binder for particleboard took over as the major adhesive in this industry. This meant that UF's were to enjoy about 90% of the adhesive field, and phenol formaldehyde had the remaining 10%. A small amount of sprayed dried UF and PF powder is also used occasionally for special products.

Particleboard came to the western hemisphere in the late 1940's, and since that time has grown with leaps and bounds. As the particleboard industry grew, so did the adhesive industry; and since the early 1950's the adhesive suppliers have kept pace with the particleboard industry and its growth supplying them with more and more sophisticated resins. The industry processes were constantly changing from the use of single opening press to the multi-opening simultaneous-closing press to caulless lines. Products ranged from homogeneous board types produced initially to the more sophisticated layered boards. General market areas were in floor underlayment, furniture core stock, panelling and in many other specialty areas. Through all of these changes and developments, the adhesive companies have tried to supply the best resin possible for each and every end use.

The original resins used on the initial homogeneous board do not in any way resemble the resin of today. These early resins were fairly simple in nature and had roughly the following physical characteristics:

Viscosity range, cps 200 - 400

pH range 7.8 - 8.2

% Total solids range 60 - 65

They had to be compatible with wax emulsions and possessed little, if any, buffer capacity. Tack requirements at that time were still undefined. Little, if anything, was known about reactivity or how to control it during these early days, and if the resin was not curing properly, an external catalyst was added.

Today, things have changed. Present day resins are more sophisticated than those used during the late 1940's. Since then, many more parameters have been added for the polymer chemists to incorporate into his new resins. We have learned the necessity for varying the formulation, altering the buffer capacity of a resin, internally catalyzing, varying the concentration, and, of course, increasing the storage stability. Control and understanding of tack is being investigated constantly. Does the resin have dry, wet, or latent tact, and, if so, to what degree? Does it retain its tack over a period of minutes or hours? Is the tack sufficient to stop edge sluffing as the mat travels from the formers to the loaders? The industry continues to require newer and faster curing resins; therefore, there is a continuing development toward this end. With the use of faster resins, precure becomes a problem; therefore, built-in safeguards must be incorporated into these resins. Lower residual formaldehyde in the board is a problem being worked on, constantly, by the entire adhesive industry. Today, resins must be compatible with the newer wax emulsions, sizing agents, fire retardants, and many other additives. The resin of today must cure at the same rate day after day regardless of the weather, wood species, or additives, in order to produce a board that is constant in its physical properties, press load after press load. And, of course, the most important thing of all, the resin must remain LOW IN COST. With all the new sophistication and background we have

gained over the years, we are still working with the same basic chemicals - namely, urea, phenol, and formaldehyde. So far, no one has found a resin system as inexpensive and as versatile as these. They now manufacture many different kinds of particleboard, from all kinds of wood species, and, types of fiber. This, of course, requires every different glues. The old fashion homogeneous board has been replaced by the layered board. Medium density fiberboard, a type of board not even thought of in the early 1940's, is now a major product. The new, continuously manufactured, thin board produced on the Bison-Mende Line has also taken its place in the market. A newer "Wafer Board" is being produced in two plants, and at least four more mills are being planned. All of these different kinds of boards require different type adhesives. The day when one adhesive satisfied many different mills is gone forever. The tack, reactivity, and viscosity of a resin used on a medium density line will certainly not work on a Mende Line. In fact, one of the most successful resins used on medium density fiberboard is the two component system. The Mende Line requires a resin of vastly different characteristics in tack, cure speed, and precure control. The Caulless particleboard line needs tack properties quite different than the conventional particleboard line. The new wafer board uses a spray dried PF resin, although some resoles and lignosulfonate resins have been tried.

A major change in species, exhibiting radically different pH buffer curves, could require a different resin. When plywood trim and other alien additives are mixed with the particleboard fibers, a new or modified resin is required. A change in end use will sometimes require a change in resins. We also are trying to introduce into these polymers many different additives for many different reasons such as fire retardants, bactericides, cure retardants, and wax emulsions. These additives are used to produce a board that will have special features that are not obtainable from fiber and resin alone. The resins of the present must have a wider range of stability

and pot life. For instance, some resins are sprayed on the chips at the blender in the form of a fine mist. Other resins might be added by droplets after the refiners or applied through the refiner shaft. Some sprayed chips leave the blender and are mat formed within a minute or two, whereas other resins are sprayed on the fiber and then held in bins for up to forty minutes before going to the formers. A change in weather, higher or lower humidity or a change in temperature, could cause a change in the tack of some resins. With all of these variables, the resin of today has to be flexible.

Because of market conditions, the board manufacturers are forced to use wood that he can obtain economically. As a result, the species may be mixed and these may vary from day to day. Be that as it may, the resin is expected to cure properly under these changing conditions and produce a board that satisfies the customer or the resin will be replaced by one that does. The rapid growth and expansion of the particleboard industry, the different type of boards produced, the different processes used, and the vastly different types of species of fiber used have put an increasing demand on the resin manufacturer to tailor-make resins for each individual plant.

In spite of our past experience and our present sophistication, there are times in the manufacture of UF and PF resins when we encounter problems in trying to make a simple change in a resin. Usually these simple changes alter so many of the resin characteristics that we essentially end up with an entirely different resin. All of the different characteristics of a resin are inter-related. TACK, DEGREE OF POLY-MERIZATION, SOLIDS, AND REACTIVITY are dependent on one another, and when one is changed, the others also change. So, for instance, if one wishes to change the solids content of a resin from 60 to 70% without changing anything else, the logical thing to do is to dehydrate the resin a little more. In doing this, however, tack and reactivity are changed. The storage stability of this resin would also be affected.

So, usually when something in a resin needs changing, it is wiser and a lot easier to develop a new resin instead of attempting to alter the old one expecting the characteristics to remain unchanged.

A friend of mine once told me that if we could invent or develop a resin that could tell the difference between solar heat, which adversely affects storage life, and platen heat, which accelerates cure, we would have it made. In many cases, and time after time, the adhesive industry has been called on to produce a resin which embodies diametrically opposed characteristics.

I have mentioned the fantastic growth of this industry several times. Well, the production figures for the year of 1973 will bear this out. The Bureau of Census reports that 3,493,648,000 sq. ft. on a 3/4" basis of particleboard were manufactured. This consumed 1,368,000,000 pounds of UF resin, on the wet basis, and 40,900,000 pounds of phenolic resin including some amount of spray dried UF and PF powder. Leonard Guss of Leonard Guss Associates, Tacoma, Washington, predicts that in 1980 there will be 9,500,000,000 sq. ft. on a 3/4" basis of particleboard produced. This would require approximately 3,705,000,000 pounds of UF resin on the wet basis. This was from an industry that started in 1946, 30 years ago.

I feel "The Future" holds many changes in store for us in this industry. I expect new and different type polymers will be tried.... ter or quatro polymers will be synthesized to fill special needs. Ter and quatro polymers are resins that have three or four monomers to be copolymerized instead of the conventional two monomers.

More natural products will be used or incorporated into polymers such as rosins, natural resins, lignin products, tannins and bark extracts to mention a few. By-products from the paper mills are sure to find a use in particleboard. By-products and still bottoms from other industries will be looked at because, in the future, current waste materials are going to disappear, both for ecological and economical reasons.

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If formaldehyde evolution from the board becomes a major problem in certain uses, then a new and different polymer or adhesive will have to be developed.

It all sounds wonderful, this unlimited growth, but the future of this industry does have a big question mark attached to it. As this industry increases in size and in resinusage, will there be enough raw materials available to fill all commitments? Almost all of the raw materials that are used in the manufacture of a particleboard resinare petroleum based. With the increase in petroleum prices and the possible shortage of this valuable raw material (OIL), it is hard to predict the future for either urea formaldehyde or phenol formaldehyde resins, let alone the future cost of the resins.

Particleboard manufacturers have in the past, and will continue to in the future, make increased demands on the adhesive manufacturer.

Sometime in the near future, I expect they will want an adhesive which, when sprayed on the fiber at 6% application rate, will produce a completely exterior board; that is, it will not be affected by heat, cold or humidity. In addition, it will not support bacteria growth or termites. It will be fireproof, and need very little, if any, finishing or painting. I imagine they will want it to be about 42 pounds in density with a 3500 MOR and 150 IB with excellent screw-holding properties. This board, of course, will have to last for years without showing any signs of deterioration or degradation.

Such a board, my friends, is the dream of every Particleboard Manufacturer, and the nightmare of every Adhesive Manufacturer.

C. H. Hickson Wood Fiber Products BORDEN CHEMICAL ADHESIVE & CHEMICAL DIV.-FAST

#### THOMAS E. BRASSELL

Tom is constantly aware of adhesives in his role as Director of Technical Services for the American Institute of Timber Construction. He originally received his Bachelor's and Master's degrees in Civil Engineering from the University of Alabama. He was Assistant Professor of Civil Engineering at the University of Alabama; Engineer with the Southern Forest Products Association; Professor of Civil Engineering at Tri-State College before joining AITC. He was Director of the AITC Inspection Bureau from 1960 to 1969, at which time he became Director of Technical Services. Tom is active in the American Society of Civil Engineers, American Society for Testing and Materials, and American Railway Engineers Association.

#### ADHESIVES FOR GLUED-LAMINATED TIMBER

Adhesives used for laminated timbers must comply with the requirements of the Voluntary Product Standard for Glued-Laminated Timber which references ASTM D 2559 for wet-use adhesives and ASTM D 3024 for dry-use adhesives. Practically all adhesives now being used are wet-use adhesives of these generic types:

Resorcinol-formaldehyde (RF), phenol resorcinol formaldehyde (PRF), melamine formaldehyde (MF), and melamine urea formaldehyde (MUF). Few, if any, casein (dry use) adhesives are being used. During the past few years lower temperature room-curing PRF adhesives have become available. Also, some manufacturers are shipping concentrated PRF adhesive resins, which require the addition of water prior to use.

For face joint bonding, room temperature or slightly elevated temperatures are used to cure the PRF adhesives, which are commonly spread on only one face with an "extruder." A few radio frequency curing beam presses are in use. Both PRF and MUF adhesives are used in these operations.

The majority of end joints used are finger joints, and MUF type adhesives are most commonly used. However, the PRF, RF, or MF adhesives must be used for preservatively treated glued-laminated timbers.

Advances have been made with proportional mixing devices using both slurry and powder for the hardening components.

The current standards for adhesives for glued-laminated timber do not limit the chemical formulations of wet-use adhesives. At the present time no new adhesives have been made available to the laminating industry as a result of these standards. The standards contain a number of performance requirements, but some additional tests need to be developed for these adhesives such as for fire resistance, impact resistance, and durability.

# ADHESIVES FOR STRUCTURAL GLUED LAMINATED TIMBERS

Structural glued laminated timbers are used as the structural framework of buildings and other structures. Therefore they must be bonded together with a strong, dependable, and durable adhesive. Adhesives used for structural glued laminated timbers should allow the full strength of the wood to be developed. As the name implies, glued laminated timber is bonded together in face joint bonding to laminate all of the plies of lumber together to form a timber that acts as a single unit. Adhesives used in this manner are stressed primarily in shear and tension perpendicular to the "glue line". The adhesives should develop approximately the same strength in shear as the wood itself. Criteria for wood failure along the shear surfaces of test specimens is usually 70% or more. Criteria for strength is 90% of the shear strength of clear wood of the species being tested. The tension perpendicular to grain strength of wood is low compared to its other strength properties. However, some individual pieces possess considerable strength perpendicular to grain and when a member is subjected to wetting and drying a high tension perpendicular to grain stress may exist and the adhesive must be capable of resisting this stress in tension.

In addition to face joint bonding, individual pieces of lumber must also be joined end to end to form full length laminations. As a general rule, adhesives are not strong in tension and cannot develop the strength of wood in tension parallel to grain. For this and other reasons it is not currently possible to join pieces of lumber end to end to form long laminations by butt jointing the pieces and gluing together. Splices must be made by cutting the wood to form a scarf joint either as a plain scarf joint or as a finger scarf joint. In either case the joint is cut so that the adhesive is stressed primarily in shear along the sloping cut. Data indicates that the flatter the slope of this cut, the stronger the joint. Slopes of from 1:8 to 1:12 have been used for plain scarf joints and slopes of approximately 1:11 or 1:12 are commonly used for finger joints. Although most adhesives can be used for either face joint bonding or end joint bonding, different curing methods frequently require special adhesives for end joint bonding.

In addition to developing the strength requirements, the adhesive must be able to function properly under the service conditions encountered. These conditions are designated as dry-use for use in covered structures where the equilibrium moisture content is 16% or less and wet-use where the member is used in an environment where the equilibrium moisture content is more than 16% such as in an unprotected location outdoors, in a moist interior environment or in marine use. At the present time very little, if any, dry-use adhesives are being used and the industry policy is to recommend that only wet-use adhesives be used for laminated timber.

Dry-use adhesives have been limited to casein adhesives and casein mixed with blood. Some urea resins were used during the late forsies and early fifties but these were discontinued because of problems with high temperature and humidity which would cause delaminations under conditions where casein was relatively unaffected.

In addition to these properties, a laminating adhesive has to be resistant to impact, fatigue, extremes of temperature including sub-zero temperatures as well

as extreme heat such as encountered in a fire situation. They must be capable of functioning in the environment to which the structure may be subjected including inorganic and organic chemicals as well as the enzyme action of mold or decay producing organisms.

The first standards for laminating adhesives were developed as military standards to meet the requirements of the armed services during and immediately after World War II. These military specifications were MMM-A-125 for casein adhesives, MIL-A-397B for wet-use adhesives with low and intermediate curing temperatures and MIL-A-5534A for high temperature curing adhesives. These adhesive specifications were referenced by the regional laminating specifications which were developed by the lumber grading rules writing agencies beginning in the early fifties. For a time the military specification CG-456 for urea adhesives was also included but was dropped for reasons mentioned earlier. These military specifications continued in use throughout the fifties and sixties. They were also incorporated as references in the Commercial Standard CS 253-63 for glued laminated timber which remained in effect until 1973. These specifications were good specifications as evidenced by their long use. From an industry standpoint, however, they had several drawbacks.

- 1. The specifications were developed for military procurement and some of the requirements were not applicable to commercial use. In most cases only the performance requirements of these specifications were adopted as references.
- The specifications contained requirements limiting adhesives to specific chemical formulations
  - a) MMM-A-125 limited the adhesive to caseins.
  - b) MIL-A-397B limited the adhesives to phenols, phenol-resorcinols, resorcinols and melamine types of adhesives.
  - c) MIL-A-5534A limited the adhesive formulations the same as MIL-A-397B.
  - d) These specifications were set up based primarily on performance with hardwoods and tests were usually conducted on maple.

These specifications contained a number of safeguards which were built in since they limited adhesive formulations to only a few chemical formulations which had a large background of testing and experience to rely on. For instance, the vacuum pressure drying test (ASTM D 1101-59) commonly called cyclic delamination test by the industry, was based on correlation of this test method to actual exposure and experience in using these adhesives.

Both the laminating industry and the adhesive industry recognized the need for adhesive specifications which were performance oriented. ASTM Committee D 14 on Adhesives was asked to develop specifications for this purpose. The task was not easy since adequate test methods for evaluating durability of adhesives as well as creep, fire resistance, etc., did not exist.

In 1970 the American Society for Testing and Materials (ASTM) published ASTM D-5525-70 for wet use adhesives and in 1972 published ASTM D-3024-72 for dry-use adhesives. These specifications differed mainly from the military specifications as follows:

- 1. The format was better suited for commercial use.
- 2. The requirement for specific chemical formulations was dropped in the ASTM specifications except for the requirement of casein in the dry-use specification ASTM D-3024-72.
- 3. Qualifying tests were to be performed on the species to be used.
- 4. Additional test requirements were added:
  - a) A steam cycle was added in addition to the vacuum, pressure, drying cycle for wet use adhesives.
  - b) A creep test was added for the wet use adhesives.

These ASTM specifications were adopted by reference in the Voluntary Product Standard for Structural Glued Laminated Timber, PS 56-73. As of this date no new chemical formulations have been proposed for use under PS 56-73. The ASTM specifications are good specifications, but they still are deficient in regard to criteria for impact resistance, fatigue, and fire resistance. Correlation of existing durability tests to new chemical formulations also poses additional questions.

At the present time practically all laminating plants are using wet-use adhesives. The majority of laminating plants use a phenol-resorcinol-formaldehyde (PRF) adhesive for face joint bonding. Several plants with large radio frequency (RF) curing presses use melamine-formaldehyde (MF) or melamine-urea-formaldehyde (MUF). A small amount of resorcinol adhesives are also used primarily for end joint bonding. The melamine urea adhesives are limited to use with untreated lumber only by PS 56-73. Existing data on melamine ureas indicate that they have somewhat less durability than other wet-use adhesives. By limiting the use of melamine urea to untreated woods, a restraint is thus placed on its use in wet locations. The phenol-resorcinol adhesives are essentially the same adhesives that have been used for a number of years but improvements in the formulations have resulted in adhesives capable of being effectively cured at room temperature or slightly below. Some adhesive manufacturers have claimed adequate cure can be obtained in the 60° to 70° range. Heat for curing is obtained by one or a combination of several methods:

- 1. Keeping the gluing room at the proper temperature.
- 2. Pre-heating cold lumber to room temperature or slightly above.
- 3. Use of heating units to heat the laminated assemblies during the curing period.

The choise of method depends on the ambient conditions existing at a given plant as well as the time available for curing.

Pressure. A minimum pressure of 100 psi is required by PS 56-73 for softwoods. Most plants use some form of a mechanical screw such as bolts or jacks to apply this pressure. A few plants use hydraulic pressure.

As a usual practice, laminated timbers are placed in a form and held under pressure over night. When curing conditions are marginal, it may be necessary to keep curved members in the forms longer than required for straight members.

Plants with the radio frequency curing presses generally use the melamine urea adhesives which must be heated to around 200° for curing. Several plants with RF presses use the phenol resorcinol adhesives. In both cases the curing time is only a matter of minutes. The exact time depends upon the size of the radio frequency generating unit and the size of the member.

Wet use adhesives of the phenol resorcinol type are generally furnished as a thick syrupy resin with a powdered hardening component which contains paraformaldehyde and a filler material. Each batch of adhesive including both the resin and the hardening component must contain a lot number. Product Standard PS 56-73 requires that each batch be tested by the block shear test and cyclic delamination test prior to using. The components are proportioned by weight. A common proportion is 5 parts of resin to one part of powdered hardener. In order to save shipping weight some adhesives have concentrated resins which require the addition of water during the mixing. Up to 20% water can be added.

Mixing is usually very simple. The mixing device should be able to distribute the powder uniformly throughout the liquid. When proportional mixing valves are used, the powdered hardening components are usually mixed as a slurry prior to piping to the mixing valve. Some automatic mixing equipment which can mix the powder with the resin is under development. The melamines and melamine ureas used are generally supplied in the powdered form which is mixed with water.

For many years the roller system of spreading adhesives on both sides of the face of a lamination was the only system used. This has been supplanted in most plants by the extruder system. The extruder system consists of a pump, a rate of flow regulating system and an extruder nozzle. The extruder nozzle is essentially a pipe with closely spaced holes. It is placed transversely over a moving lamination. Multiple small streams of adhesive flow through the holes in the nozzle and are deposited on the lamination as closely spaced strips of adhesive with a cross section that is approximately semi-circular. The advantages of the extruder are:

- 1. It permits adhesive to be spread on only one side which allows laminations to be transported on rollers without wiping off the adhesive.
- 2. Because less surface area of the adhesive is exposed, the assembly time is generally longer than with roller spread adhesives of the same spread rate.
- 3. The rate of spread is usually easier to control.
- 4. Because assembly times are usually longer than roller spread adhesives, it is often possible to use a lower spread for a given ambient condition.

In most instances adhesives are mixed in batches and stored in a reservoir on the glue spreader. Some waste is inevitable since it is very difficult to mix the exact amount required for a given job. The more commonly used extruder has a nozzle longer than the lamination is wide. To insure full coverage of the lamination, the excess streams of adhesive that fall past the lamination are collected in the reservoir and recirculated. The energy imparted to the adhesive during the pumping required for recirculation will lower the pot life of the adhesive. For this reason most plants cool the resin prior to using in order to keep the temperature in the reservoir within an acceptable range. A few plants circulate cool water in a jacket around the adhesive reservoir.

All adhesive mixing equipment has to be cleaned frequently. One of the required properties of a laminating adhesive is that it will allow mixing and spreading equipment to be easily cleaned. Disposal of the waste water used in cleaning sometimes poses a problem. Several methods are being investigated to reduce or eliminate this problem.

- 1. Proportional Mixing. A proportional mixing valve can be located near the nozzle and only a small amount of water is needed to clean the spreader. In this case a nozzle the exact width of the lamination must be used so that all of the adhesive falls on the lamination and none of the adhesive is recirculated. Also extremely accurate sensing devices and fast acting valves are needed to prevent waste at the ends of the lamination.
- 2. Recycling of waste water. In adhesive systems where the resin is shipped in a concentrated form, and water is to be added at the plant to bring the adhesive up to the proper viscosity, the water used for this purpose can be the wash water which was previously used to clean the spreader and mixer: Care must be used to prevent this wash water from becoming contaminated with grease or other undesirable foreign matter.

The spreading of adhesives on end joints is accomplished in a number of ways ranging from hand spreading to use of disc type automatic spreaders. The adhesive may be spread on one end only or on both ends of the finger joint. Most finger joints are cured in RF presses. The melamine-urea is frequently used for end joints because of its adaptability to automatic equipment, ease of cleaning and ease of curing by radio frequency. Its electrical properties do not induce arcing as much as may occur in some other adhesives. A few plants use phenol resorcinol for end joints cured by radio frequency or in some cases the joints are cured at room temperature when the production is small.

Gluing of treated lumber poses no special problems. Most adhesives commonly used for untreated lumber can be used to glue treated lumber. PS 56-73 requires that each species-treatment-adhesive combination be qualified prior to using. Although it is technically feasible to glue most preservatively treated lumber, current practice is to glue only the lumber treated with the following types of preservatives:

- 1. Pentachlorophenol in liquid petroleum gas or a light solvent.
- 2. Water borne salts such as CCA or ACA.

Lumber treated with pentachlorophenol can be glued almost as easily as untreated wood. Lumber treated with water borne salts is somewhat more difficult to glue. In all cases, however, the lumber must be resurfaced after treating shortly before gluing. It is very important that all surfaces be planed clean of preservatives prior to gluing.

Lumber treated with fire retardant is difficult to glve and usually requires a specially compounded adhesive for this purpose.

#### New Developments

Most adhesives now being used are of the same generic types that have been used for many years. All of these adhesives are based on a thin glue line since these

adhesives are non-gap-filling. Recently a new gap-filling phenol-resorcinol has been developed which will allow the gluing of less precisely surfaced lumber and will permit the gluing together of large sections of laminated timber for such purposes as making I shaped sections out of smaller members.

No other new adhesives have reached the stage of development where they are ready for use in laminating in this country.

## Summary

In conclusion, the adhesives used for laminated timbers are of the same generic types that have been proved by years of experience. These are phenol-resorcinol, resorcinol, melamine urea and melamine. The currently used ASTM specification for wet-use adhesives will permit adhesives of other chemical formulations to be used. These new specifications have been in effect only since 1973 and it is too soon to predict at this time what effect they will have on development of newer adhesives.

#### ROBERT H. GILLESPIE

Bob is in charge of research on adhesive systems and adhesive-bonded products at the Forest Products Laboratory. He received his B.S. degree in chemistry from Indiana University in 1938, and Ph. D. in organic chemistry from the University of Wisconsin in 1944. He spent 15 years in research and development in the textile and adhesive fields in New England, in the Boston area. He was on the teaching staff and conducted research at the Institute of Paper Chemistry in Appleton, Wis., for 2 years before coming to the Forest Products Laboratory in 1960. In 1965 he moved up to his present position.

## ADHESIVES FOR HOUSING, A WORKSHOP REPORT

The Forest Products Laboratory sponsored a workshop in April 1975 on Adhesives in Building Construction. About 25 people attended, representing universities, Government laboratories, manufacturers of adhesive ingredients, adhesive formulators, homebuilders, regulatory agencies, and quality control and inspection services. The objectives of this workshop were twofold—first, to define the constraints that prevent widespread use of construction adhesives for building assembly, and second, to develop plans to realize the full potential of construction adhesives.

The emphasis was on construction adhesives of the low-modulus type, formulated with elastomeric bases. The whole gamut of problems were discussed—for instance, those associated with adhesive use in building construction, potential new applications, deterrents to acceptance and use of new applications, designing structures with adhesives, evaluating adhesive properties and performance, research opportunities, and methods for improving communications between various interests involved in the housing industry. The workshop stimulated some of the attendees to write specifications for submittal to FHA and ASTM. Others want to plan future sessions to explore suggested new uses for adhesives in building construction in greater depth.

# ADHESIVES FOR HOUSING--A WORKSHOP REPORT

### Introduction

This is a very brief summary of a workshop on construction adhesives that was held at the Forest Products Laboratory this past April. The objectives of this workshop were twofold: First, to define the things that interfere with using construction adhesives in building assemblies. Second, to plan how we might realize the full potential of construction adhesives. The emphasis was on construction adhesives, especially of the low-modulus type formulated from elastomeric materials.

These nonrigid adhesives gained acceptance by the building industry through the development of the field-glued floor systems. The adhesives are available in cartridges and are easily extruded by caulking guns. Usually the operator lays a bead along the top of the joists, and in the joints between plywood sheets as they are installed on the joists. With the adhesive, only half the usual number of nails are required. The nails hold the plywood in place as the adhesive develops its full strength. By bonding the plywood at the joints, and then to the joists, a diaphragm T-beam structure is produced. Such a diaphragm has greater stiffness than unbonded floors. Lighter joists for a given span can sometimes be used. Consequently, this saves building material. An added benefit was the elimination of "squeaky" floors. The success of these mastic-type construction adhesives in floor systems has led to other applications in building construction.

### Attendees:

The people attending the workshop had a wide divergence of viewpoints. However, all shared a common interest in the building industry. To show you the quality of people, those from universities included Bob Hoyle of Washington State, Gordon Krueger from Michigan Technological University, Stan Suddarth from Purdue, George Atherton and Tony Polensek from Oregon State, and Jim Koutsky from the University of Wisconsin. Giving a little different viewpoint was Don Percival of the Small Homes Council at the University of Illinois.

Representing the manufacturers of elastomeric materials or of formulated adhesives were Hal Roth and Austin Snow of Dupont, Dick Hartman of B. F. Goodrich, Bev Garrett of Roberts Consolidated Industries, and George Butts of the Franklin Glue Company.

For the Canadian Forestry Service, Donald Onysko participated.

The U.S. Forest Service was represented by Charles Vick of the Southeastern Forest Experiment Station and Henry Montrey, Mike Millett, Bryan River, and myself from the Forest Products Laboratory.

Willard Worth of the National Homes Manufacturing Company provided provided the viewpoint of a progressive home manufacturer. Ed Starostovic of the Product Fabrication Service provided information on home manufacturing, inspection and quality control, and code and regulatory problems. The viewpoint of our Federal regulatory agency was supplied by Howard Hilbrand of the Department of Housing and Urban Development of the Federal Housing Administration. Finally, Ed King of the National Forest Products Association, represented the manufacturers of basic building materials. All in all, the attendees brought together a vast background and experience in the housing industry.

## Technology Distribution in the Building Industry

The complex nature of the building industry leads to many constraints to more widespread use of adhesives in building construction. How can new technology be introduced and new products accepted with confidence? This subject was discussed at length. First discussions were by homebuilders and developers who supply houses for the housing markets. They build on-site, provide components for field assembly, or supply factory-built homes in panelized, modular, or mobile home form. Such homebuilders and developers purchase raw materials and equipment from many different manufacturers. These materials and their assembly must meet the codes of the local regulatory agency. Technical information is often supplied by architects and engineering companies. For all of this to work requires capital. Because they most often supply that capital, lending institutions exert considerable influence just by their willingness to make loans. The research and development performers, whether they be universities, Government laboratories, trade associations, industrial laboratories, or consultants are faced with a bewildering maze. They must reach a broad array of companies and organizations to pave the way for new technology. Only then can use and acceptance of the new product or process be expected. The R&D performer is involved with code and regulatory agencies, architect and engineering companies, equipment and basic material manufacturers, homebuilders and developers, and even lending institutions, to apply new technology successfully in the home-building industry.

The viewpoint of code and regulatory agencies is crucial. In fact, use and acceptance of new adhesive technology in the building industry is like working in a jungle. Who writes new specifications, test methods, and who develops new standares? Who sees that these are then incorporated in building codes and regulations?

There are many organizations to consider and many directions in which to go. But there is no clear path to follow in this area. The problem is further complicated because the expertise in adhesive technology is widely diffused. Many interrelated industries have people with abilities, but without a central focus on assessing problems and initiating action. Probably the most inclusive membership on adhesive expertise is that of ASTM Committee D-14 on Adhesives. However, the scope of action of this committee is limited. Experience has shown that good standard development requires much time, much patience, and much compromise.

## New Ways of Using Construction Adhesives

At one point during the workshop, a small group split off by themselves in a brainstorming session. This was referred to as an experiment in "Synectics." Its purpose was to uncover new ways of using construction adhesives in buildings. I am not going to present the results of this think-tank. Later on John Rose will discuss some similar information. This will be in his talk on construction adhesives. I think the brainstorming group felt the session worthwhile. They have already planned another meeting to consider a "wind-resistant" house. This would be one that would survive tornadoes or hurricanes better than conventional structures. Naturally, construction adhesives and new design concepts would be an integral part.

# Panels for House Assembly

When stressed-skin and sandwich panels were first developed, a few housebuilders tried to use them as factory-built panels, with later assembly on-site. There were two attractions. One was a possible saving

in materials. The second was that space would be saved in transporting the panels to the job site. Thin panels would pack more efficiently in the available truck space. However, thin panels, bonded with rigid glues, often warped with changes in moisture conditions. The anticipated advantages of these bonded panels could not always be realized.

We now recognize that thin lightweight panels do not answer all problems. Today we expect adequate strength and stiffness from a panel, but also satisfactory performance acoustically, thermally, and against fire. To explain, some attention must be given to reducing sound transmission. There must be enough space in outside walls for insulation and to pass fire performance tests. These considerations have led to thicker panel sections with specific designs where adhesives can play a definite role.

# Engineering Designs

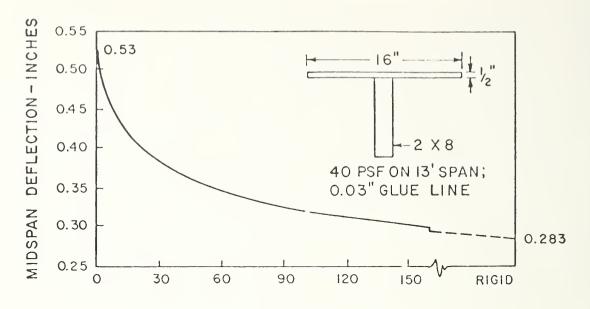
The major question with nonrigid adhesives has been whether or not they have a place in structural or semistructural applications. In such applications, the bonding contributes to the strength and stiffness of a structure. This allows less material to be used, but still perform adequately.

The first demonstration that non-rigid adhesives were useful in structural applications was by Kuenzi and Wilkinson of the FPL. They theorized that a nonrigid adhesive provided partial interaction between laminae in a beam. A rigid adhesive, on the other hand, gives complete interaction between the laminae. There is no interaction when the laminae are not bonded. The amount of interaction provided by the nonrigid glueline was dependent upon the shear modulus of the adhesive and the thickness of the bond line.

A similar approach was taken by Bob Hoyle at Washington State. He calculated the structural performance that would be expected with adhesives of different shear moduli. Figure 1 shows what he is finding. This is a plot of how much a T-beam would deflect under a load of 40 pounds per square foot, as the modulus of rigidity or (G) value of the adhesive was varied. For his T-beam, Bob used a 2 x 8 bonded to 1/2-inch plywood 16 inches wide. This was set up on a 13-foot span. He assumed a glueline thickness of 30 thousandths of an inch. Under this 40-pound-per-square-foot load, an unbonded beam would deflect 530 thousandths of an inch.

You can see how rapidly this deflection is reduced by adhesive bonding. A modulus of rigidity of only 90-100 has reduced deflection almost as much as when a rigid adhesive is used. Hoyle has many other examples which show much the same thing--that adhesives with shear moduli between 50 and 100 p.s.i. can perform almost as well as a rigid adhesive. Results of this type get us all excited about using low-modulus adhesives in building construction.

The design engineers in the group discussed methods to predict the performance of structures. These included the conventional methods, where laboratory measurements are converted into design values. Of course, this conversion is by factors that account for variability, exposure conditions, quality control, duration of load, and safety. But engineers are also bringing more sophisticated methods into use. George Atherton and Tony Polensek talked about their work with finite element analysis and how it could be applied to bonded structures. Mention was also made



MODULUS OF RIGIDITY OF ADHESIVE, G - PSI

Figure 1

	SHEAR MODULUS (psi)		SHEAR STRENGTH (psi)
ONE PART POLYURETHANE	30-150	WIDE RANGE OF TEMPERATURE, MOISTURE, AND STRESS LEVEL	100-350
NEOPRENE-RESIN MASTIC	50-5,100		40-900
SITKA SPRUCE (ROLLING SHEAR)	3,500-55,000		
1/4" HARDBOARD	75,000		500

Figure 2

of current methods that are called in some quarters as "limit state" design which is based upon probability assessments or reliability analysis.

## Properties of Adhesives Needed for Engineering Design

All of these methods for engineering design require more information about the properties of the adhesives being used. The questions are "What kind of information?" and "How should the properties be measured?"

The most important information required is on shear strength, shear modulus, creep in shear, and duration of load. In special cases, it would be desirable to have information about strength in tension, cleavage, or peel. I will confine my remarks to shear properties only. The reason, of course, is that the most satisfactory designs will place bondlines so they will be stressed predominantly in shear.

Methods for measuring shear strength, shear modulus, creep in shear, and duration of load are currently being developed by Krueger at Michigan Tech, and River at the Forest Products Laboratory.

Gordon Krueger has developed a testing device with great promise.

He has tried to combine the best features of a torsion test with those of a "rail test." In a rail test, a thin specimen is rigidly clamped on each side of the bondline. The assembly is then compressed in a testing machine in a skew fashion. Thus the adhesive receives a shear stress as well as a small normal component of stress. Krueger converted this rail test into a torsion test by bending and clamping his specimen to the peripheral surfaces of two large-diameter wheels. The thin specimens can be rigidly supported with relatively stiff clamps. This system produces a uniform shear stress distribution in the longitudinal direction of the bondline without the normal stress component. The apparatus is

particularly useful in measuring the shear modulus of low-modulus adhesives

River has developed specimens that are essentially thick parallel plates stressed in tension. With these, he measures short-term load-deformation properties such as shear modulus and shear stress. He also uses it for long-term load-deformation properties associated with creep and rupture stress under long-term loads. Each method has its own set of advantages and disadvantages. Further research and use of the methods is needed before we know which might be preferred.

Two major problems face the developers of these test methods:

One is that properties must be measured under a wide range of conditions.

The other deals with judgment factors in interpreting the measurements and using the data judiciously in engineering design.

With regard to evironmental conditions, low-modulus adhesives are sensitive to temperature, moisture conditions, stress levels, and duration of load. Each formulation responds in a different way to these influences. It is not like dealing with wood, whose shear strength and modulus changes little with the temperature differences found in houses. Design engineers have a number of adjustment factors they use when designing with wood. Experience has shown they work quite well, although wood scientists continue to search for better precision in these adjustment factors. But measuring the mechanical properties of low-modulus adhesives is different. Such measurements must be made under different conditions of temperature, moisture content, load, and time before their behavior can be adequately described.

The uncertainty about interpreting the results and using the data wisely is due to the visco-elastic character of the low-modulus adhesives.

This is particularly a problem with shear modulus. I might remind you that this term of shear modulus has a lot of meaning and usefulness to design engineers. It is a measure of a material's resistance to deformation when a small force is applied to it. By definition, it is the ratio of shear stress to shear strain. Now this ratio is a constant when measured properly—and this means at strains less than the elastic limit.

It is true that material is perfectly elastic. However, in a practical sense, most materials of construction are somewhat elastic.

Thus a value for shear modulus is essentially constant and is meaningful.

But some elastomeric adhesives deform in a way where it is all but impossible to find an elastic limit. In such cases, the shear modulus is not a constant since some plastic deformation takes place even at low strains. In these cases, it is not completely correct to call this measurement a shear modulus. But I'm sure shear modulus values will continue to be reported for construction adhesives to describe resistance to short-term loads and small deformations.

Here we have a neoprene-based adhesive and a polyurethane adhesive. Note that modulus and shear strength vary widely depending on the temperature, moisture, and stress levels, and other conditions of test. You can compare these shear modulus values with Bob Hoyle's illustration.

Remember that a shear modulus of 50 p.s.i. or greater provides benefit. Thus you'd conclude that either of these adhesives might be used in construction forcertainservice conditions. As a matter of comparison, I have included shear modulus values for Sitka spruce in rolling shear and for hardboard. These values were obtained by the same test procedure and equipment at ambient room temperature and moisture.

#### Other Constraints

So far I have talked mostly about technical constraints to more widespread use of construction adhesives in building. These restraints include the need for test methods, collection and interpretation of data on their mechanical properties, and the need for cooperation and understanding between everyone. But there are nontechnical constraints also. That array could be discouraging if we'd let it. For example:

- 1. We need specifications a builder can relate to and understand.
- 2. Builders need to know the benefits of adhesive bonding, and the alternatives in adhesive selection and use.
- 3. We need data sheets on adhesives that a builder can understand.
- 4. Some builders are reluctant to use adhesive systems that are more costly and difficult to use than the familiar white glues or PVA's.
- 5. Construction adhesives take too long to reach full strength after bonding.

The list can go on and on, but none of these constraints appear insurmountable.

To solve them involves developing good systems of technology distribution,

appropriate specifications, and programs to put factual information into

the right hands.

Most attendees felt a need for classifying construction adhesives into classes. The class would depend on adhesive properties and be related in turn to applications. One suggestion proposed three classes such as A, structural; B, Temporary or Semistructural; and C, Utility or Cosmetic.

#### In Summary

I feel the objectives of the workshop were met. We identified many constraints to increased use of construction adhesives. A few new applications were proposed. The R&D performers went away with a clearer idea of the direction their research should take. One attendee volunteered to rewrite Krueger's torsion rail test in the ASTM format and send it on its way toward standardization. Another offered to put together some test method proposals for a specification of adhesives for use in mobile homes. The brain-storming group planned future meetings. So, some positive action resulted from the deliberations.

This workshop helped to unravel the complex problems associated with expanding the use of construction adhesives in buildings. The information exchange gave each one attending a better understanding of the interactions that must take place before new technology can be put into successful practice. No doubt other workshops on the same subject will follow.

### ROBERT F. SNIDER

Bob is Sales Manager of the Industrial Division of the Franklin Glue Company. Woodworking adhesives have been a big part of his 39 years with Franklin--years in which his activities ranged from research to production and sales. He has shared the resulting information in many articles on the use of adhesives in the woodworking industry. His formal training was at Ohio State University, culminating in a Ph. D. degree in chemical engineering.

#### ADHESIVES IN FURNITURE JOINTS

Furniture quality depends on the quality of the glue joints. To get glue joint quality, good joint design is important. Glue joints should be made to maximize side-to-side grain contact. Joints such as dowel, tongue and groove, mortise and tenon, and dovetail are all used to this end. Improper selection or poor design of the joint can result in joint collapse, poor appearance, veneer rupture, telegraphing, etc. Moisture content should be chosen to fit the conditions of field exposure. Wood species will affect the stability of the piece when subjected to stress and will also determine the stresses introduced by moisture change. Manufacturing variables such as surface preparation, fit, glue application, amount of pressure, and pressure time will affect joint quality.

The surface to be glued should not be covered with loose fibers nor should it be glazed. Fit should be snug. Dowels and tenons should be coated as well as the holes and mortises to insure that too much glue has not been rubbed off or pushed to the bottom of the hole. Pressure for both edge and laminating gluing should be adequate to bring the joint tightly together but not enough to crush the wood. Pressure is required to hold the joint tightly together until the adhesive has set adequately so that absolutely NO springback occurs.

Although there are adhesives which will perform well for almost all wood-working uses, specialized adhesives developed for specific operations and conditions will perform better than the ones developed to perform the gamut of operations.

The expertise of the adhesive manufacturer can be helpful in the selection of the appropriate adhesive and advising on its proper use.

Information is based heavily on the publication "Gluing and Furniture Design" from the Franklin Glue Company, Columbus, Ohio 43207.

#### ALAN A. MARRA

Alan is professor of Wood Science and Technology at the University of Massachusetts, College of Food and Natural Resources, Department of Forestry and Wildlife Management. With three degrees in the field of Wood Science and Technology (BS and MS, N.Y. State College of Forestry; Ph.D. University of Michigan), he has specialized in adhesives, wood modification, and wood composites during 33 years of teaching, research and industrial practice. He also holds patents and patent applications in wood composite materials and processes.

Currently he is engaged in research on wood-foam composites as a means of converting low-grade wood into high-grade products with low-grade funding.

# ADHESIVES IN COMPOSITES

An analysis of the processes by which two pieces of wood are bonded together produces a line of reasoning which relates most glued products in terms of origin and quality of the wood, its breakdown into primary elements, reorganization, and reconsolidation into secondary products of prescribed properties. The role of the adhesive is seen not only as a bonding agent but also as the chief determinant of efficiency in manufacturing the products.

Developed in the context of conventional products and processes, the reasoning is extrapolated to new situations and used to draw a set of specifications for adhesives not yet in existence. The new situations include products which are thicker than are normally produced, products which are lower in density, and products which represent greater formability.

In addition to the conventional requirements of high durability and low cost, the desired new adhesives should be fast curing, preferably without the need for high-energy input in the form of heat and pressure. In addition the adhesives should be easy to apply uniformly to the wood. This may be more of an equipment problem than a chemical problem but elements of both will influence the solution.

Based largely on information from: "Low Density Composites from High Density Hardwoods," by Alan A. Marra, Wayne A. Hausknecht, and Robert F. Day. It was published February 1975 by the Massachusetts Agricultural Experiment Station as Bulletin No. 610.

#### W. W. MCCANN

"W.W." is Manager of Research and Development for the Simpson Timber Company. He is a graduate of Washington State University, where he majored in Chemical Engineering. During his military experience, he spent 2-1/2 years at Los Alamos, N.M. on the Manhattan Project. His work has also included 4 years as a mill chemist at a pulp mill for Rayonier Incorporated, 2 years at Simpson in research and development and 14 years as production superintendent in insulation board manufacture. For the last 13 years he has been back in R&D, as Commercialization Manager and in his present capacity.

# ADHESIVES FOR OVERLAYS AND PATCHING

- 1. Overlays and Overlaid Products:
  - a. Brief description of types and uses.
  - b. Brief description of manufacturing processes and application.
  - c. Performance aspects and critical factors.
- 2. Veneer Patching Methods in Use:
  - a. Types and application.
  - b. Problems and opportunities.
- 3. Future Needs:

# ADHESIVES FROM NONPETROLEUM RESOURCES

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#### K. C. SHEN

Dr. Shen is a research scientist at the Eastern Canadian Forest Products Laboratory at Ottawa. He is a native of China and received his Bachelor's degree there before acquiring his M.F. at the University of British Columbia and Ph. D. at the University of Tokyo. His professional experience includes work at the Western FPL at Vancouver as a research assistant; as a research engineer at Abitibi Paper Co, in Toronto; and as production supervisor at Wigewood Waferboard Ltd. of Saskatchewan before he moved to his present position.

# BONDING CHARACTERISTICS OF ACIDIFIED AND SPRAY-DRIED SPENT SULFITE LIQUOR

We have developed recently at the Eastern Forest Products Laboratory a system utilizing spent sulfite liquor (SSL) as a binder for making exterior-grade waferboard. This system involves, first, acidification of concentrated SSL with sulfuric acid and, second, spray-drying the mixture under controlled conditions to give a powdered resin. This powder is dark, highly hygroscopic, and water-soluble. Preliminary work indicates the acidified, spray-dried resin powder is stable and produces satisfactory bonds after almost 2 years of storage in dry conditions.

Exterior-grade poplar waferboard blended with a 4-5% content of SSL powder can be made, using a press condition very close to that of phenolic waferboard, to meet the physical and strength properties specified by Canadian Standards Association 0188 (68).

This report describes our current work on the SSL-powder system. It also presents basic information on the bonding characteristics of the resin as affected by its acid level, resin content, press temperature, and time.

Two hundred and sixteen 2-ply poplar veneer panels  $12- \times 12- \times 1/4$  inch  $(30.5 \times 30.5 \times 0.63$  cm), were assembled using SSL powder containing 4 levels of acidity, three loadings of resin, and pressed at three temperatures, for three different times. Specimens obtained from these panels were aged according to the standard and new accelerated treatments prior to tensile-shear test. Such accelerated aging treatments included vacuum pressure soaking, cyclic boiling, ASTM D 1037, dry heating at 300° F (150° C), and closed wet heating at 250° F (120° C), 10% moisture content.

Test data from these experiments will be summarized and results discussed at meetings.

#### ARTHUR B. ANDERSON

Now officially retired, Art carries on his long-standing interest in wood research even though the records say he is "Biochemist Emeritus of the Forest Products Laboratory at the University of California." Art received his BS at the University of Wisconsin in 1929 and his Ph. D. in 1933, and was a research assistant at the U.S. Forest Products Laboratory from 1929 to 1934. He spent 6 years with Quaker Oats Company as a research chemist, was chief chemist for the Western Pine Association for 8 years, and director of research for the Oregon Lumber Company for 2 years. From 1950 to 1974 he was a biochemist at the California FPL. His sabbaticals were spent at the Norwegian Pulp and Paper Institute in Oslo, the Philippine Forest Products Research Institute, and most recently at New Zealand Forest Products Ltd. in Auckland.

# BARK EXTRACTS AS BONDING AGENT FOR PARTICLEBOARD

Future growth and development of the particleboard industry may depend largely on sources of phenolic adhesives other than petroleum. Further, it would be desirable that a low-cost raw material be a renewable source of phenolics.

One of the ubiquitous by-products of the forest products industry at plant site is bark residues. A recent survey in Oregon indicated that bark was the least utilized of all types of residues, and bark disposition is becoming more of a problem due to increased restrictions on the incineration of bark residues.

Chemical processing of barks is limited, and the principal chemical products produced commercially from them are based on bark's phenolic content. Barks generally are much richer than wood in quantity and complexity of extractive components, the most important being monomeric polyphenols or flavonoid compounds, and polymeric phenolics such as tannin, phlobaphenes and phenolic acids.

Use of phenolic components of bark extracts in preparing adhesive components used in plywood and particleboard manufacture has been proposed from time to time. Such preparations are based on the reaction of bark phenolic components with an aldehyde, usually formaldehyde.

This discusses an investigation undertaken to evaluate the bark extracts from four west coast species, including white fir, ponderosa pine, Douglas-fir and western hemlock as bonding agents for particleboard.

Dr. Anderson's talk was based largely on the following publications:

Anderson, A. B., A. Wong, and K. T. Wu

Utilization of White Fir Bark in Particleboard For. Prod. J. 24, No. 1, p. 51, 1974

Utilization of White Fir Bark and Its Extrict in Particleboard For. Prod. J. 24, No. 7, p. 40, 1974

Utilization of Ponderosa Pine Bark and Its Extract in Particleboard For. Prod. J. 24, No. 8, p. 48, 1974

Douglas-Fir and Western Hemlock Bark Extracts as Bonding Agents for Particleboard For. Prod. J.  $\underline{25}$ , No. 3, p. 45, 1975

#### H. M. SAAYMAN

Dr. H. M. Saayman is Chief Research Officer and Head of the Wattle Research Section of the Leather Industries Research Institute, Grahamstown, Republic of South Africa. He received the M. Sc. degree in Physical Chemistry at Rhodes University, Grahamstown, in 1962 and his Ph. D. in flavanoid tannin chemistry in 1966. He has been a member of the staff of the Leather Industries Research Institute for 13 years. During 1969 - 1970 he spent a year doing post-doctoral research work on the chemistry of catechins at the Institute of Organic Chemistry of Heidelberg University, West Germany.

#### WATTLE TANNIN ADHESIVES FOR WOOD BONDING

Black wattle bark tannins have been used in the production of a wide range of adhesives for products from wood.

Thermosetting tannin-formaldehyde adhesive resins have been developed for use in the manufacture of particleboard, plywood, and blockboard. In addition, a wattle-based cold-setting adhesive formulation has been produced for the lamination of wood.

Unfortified adhesive resins, prepared from wattle tannins, have been used to manufacture exterior-grade particleboard. Commercial production of weatherproof board started 10 years ago in South Africa and the product has found application in the construction of freeway road signs, as outdoor cladding in prefabricated buildings, and in the general building trade.

Two different tannin-based plywood adhesives have been developed. For interior applications an unfortified tannin-formaldehyde formulation was found to give adequate bonding. In the manufacture of exterior-grade plywood, however, it was found necessary to fortify wattle-based adhesives with phenol- or resorcinol-formaldehyde resins.

Marine plywood production required the addition of a resorcinol-phenol-formaldehyde fortifying resin. For exterior-grade plywood and blockboard the wattle-based formulation was fortified with a phenol-formaldehyde resin to improve outdoor weathering properties.

A chemically modified cold-setting resin was developed for use in finger jointing of short lengths of timber and in lamination of timber beams for structural purposes. This tannin-based resin contains a substantial amount of resorcinol.

# WOOD ADHESIVES FROM WATTLE

BARK EXTRACT

by

H.M. Saayman and J.A. Oatley Leather Industries Research Institute P.O. Box 185 Grahamstown 6140 Republic of South Africa

#### SUMMARY

Black wattle bark tannins have been used in the development of a wide range of adhesives for products from wood. Thermosetting tannin-formaldehyde adhesive resins have been produced for use in the manufacture of particleboard, plywood and blockboard. In addition, a wattle based coldsetting adhesive formulation has been developed.

Unfortified tannin-formaldehyde adhesives have been used in the manufacture of exterior grade particleboard.

Two types of tannin based plywood adhesives have been produced - for interior applications an unfortified formulation gave acceptable bonding, while exterior grade plywood required fortification with phenol- resorcinol-formaldehyde resins.

A wattle based formulation has been developed for use in the manufacture of blockboard or shutteringboard. This adhesive was fortified with a phenol-formaldehyde resin.

A chemically modified cold-setting resin has been produced for timber lamination and finger jointing. This tannin based resin contained a proportion of resorcinol.

The general versatility of wattle bark tannins as raw material in the production of wood adhesives is evident from these developments.

Keywords: Tannin-formaldehyde; Particleboard; Plywood; Cold lamination.

#### INTRODUCTION

The bark extract of the Black Wattle tree (Acacia mearnsii De Wild) contains approximately 70 per cent of polyphenolic tannins, as determined by the official hide powder method of tannin estimation(1). The moisture content of spray dried extract usually amounts to 5-6 per cent, while the balance of the material is composed mainly of resins derived from carbohydrates(12).

The condensed tannins are polymers of hydroxylated flavanoid constituents(5). The phenolic hydroxyl groups are highly reactive and they induce reactive centres in the ortho- and para-positions of the benzenoid nuclei as shown in Fig. 1.

centre

Fig. 1.

The nucleophilic centres on the A-ring of a flavanoid unit tend to be more reactive than those found on the B-ring. This is due to the vicinal hydroxyl substituents which merely cause general activation in the B-ring without any localized effects such as those found in the A-ring.

In their reactions with formaldehyde the flavanoid tannins behave as typical phenols. The reactivity of wattle bark tannins is somewhat closer to that of resorcinol than of phenol. Formaldehyde reacts with tannins to produce polymerisation through methylol bridge linkages at reactive positions on the flavanoid molecules.

Initial studies on the use of tannin-formaldehyde resins as adhesives for wood were conducted by Dalton(2,3). Subsequent work by Plomley and co-workers has shown that wattle bark tannins are suitable raw materials for the production of adhesives resins used in the bonding of plywood and particleboard (8,9,10).

Research work relating to wattle tannin adhesives was initiated in the Wattle Research Section of the Leather Industries Research Institute in 1964. Since then a wide range of adhesive resins has been developed for products from wood.

The present publication describes tannin-formaldehyde adhesives for bonding products such as particleboard, interior- and exterior-grade plywood, blockboard and laminated timber beams. All adhesives used in these applications are themosetting, except the last mentioned resin, which is cold-setting in character.

#### EXPERIMENTAL AND RESULTS

#### A. Particleboard

Unfortified wattle tannin solutions were catalysed with formaldehyde solution, powdered paraformaldehyde or a solution of hexamethylene tetramine to produce adhesive resins used in the manufacture of weather resistant particleboard.

In order to reduce the relatively high viscosities of tannin solutions containing greater than 45 per cent of solids, it was found necessary to subject the extract solution to an alkaline pre-treatment. This served a dual purpose - firstly, it lowered the viscosity of the tannin solution and secondly, it conferred a longer pot-life on the final tannin-formaldehyde adhesive.

The glue mix was prepared as follows:

#### Glue mix.

Constituents	Parts by Mass
Wattle extract	45
Water	55
Caustic soda (50% aqueous)	4
Glacial acetic acid	3
Formaldehyde (40% aqueous)	11,25

A solution of wattle extract, containing 45 per cent by mass of solids, was prepared by dissolving dried wattle extract powder (45 parts) in water (55 parts) and adding caustic soda solution (4 parts, 50 per cent by mass) to adjust the pH level to 9,5. The mixture was refluxed at 105°C. for 60 min. and cooled to ambient temperature. Glacial acetic acid (3 parts) was added slowly with stirring to lower the pH to 7,0. The viscosity of this solution was in the range 100-150 c.P. at 25°C.

To the extract solution was added formaldehyde solution (11,25 parts of 40 per cent by mass) and mixed thoroughly before spraying the adhesive resin on the wood chips in the blender. The effective pot-life of the resin was 3-4 hr. under these conditions and 10 per cent of adhesive was used on the mass of dry chips.

In addition to the resin a wax emulsion was sprayed separately on the wood chips, after the adhesive had been applied. The emulsion, "Mobil ED 80/53" contained 66 per cent of wax solids and was applied to the wood chips on a basis of 1 per cent by mass.

Twenty particleboards of average thickness 12-13 mm. were manufactured according to the Bähre-Bison system with the above tannin-formaldehyde adhesive resin. Boards made with this system consist of a central layer of coarse wood chips while graded layers of finer chips form the outer surfaces.

The boards were pressed at temperatures varying between 135° and 200°C. for times ranging from 3 to 15 min. The densities of the finished boards were chosen to be in range of  $690-710~\mathrm{kg./m.^3}$  in order to obtain values which were directly comparable with coards produced with weatherproof phenol-formaldehyde adhesive resins.

After trimming and sanding, the boards were left to post-cure for 24 hr. before being conditioned at 20°C. and a Relative Humidity of 60 per cent for 24 hr. prior to testing. The thickness of each panel was measured at several positions and samples were prepared for determination of density, bending strength (modulus of rupture), tensile strength perpendicular to the face of the panel and swelling after immersion in cold water for 2 hr. and 24 hr. periods. Duplicate samples for the tensile test were prepared - one for evaluation after soaking in cold water (20°C.) for 24 hr. (V20) while the other was tested after soaking in boiling water (100°C.) for 2 hr. (V100).

The average results of sets of five boards each are summarized in Table 1.

<u>Table 1</u>

<u>Average Results of Physical Tests Conducted on Particleboards</u>

<u>Manufactured with Tannin-Formaldehyde Adhesive</u>

Boards	Press Temp. °C.	Heating Rate sec./mm.	Press Time min.	Thickness mm.	Density kg./m.
1-5	135	74	15,0	12	700,0
6 <b>–</b> 10	170 200	30 20	6,1 4,05	12 12	691 <b>,</b> 9 690 <b>,</b> 8
16–20	200	15	3,05	12	703,3
German Specification DIN 68761 (3) (1967) (4)			8-13 (range)	750 (max.)	

Boards	Bending Strength kp./cm.²	Tensile Strength kp./cm.²		Percentage	e Swelling
		V20	V100	2 hr.	24 hr.
1-5	171,5	5 <b>,</b> 9	1,1	5,4	17,4
6–10	224,3	5,5	1,3	4,8	15,4
11–15	221,5	4,9	1,4	4,4	15,2
16-20	219,4	4,9	1,4	4,5	14,7
DIN 68761 (3) (1967)	200 (min.)	4,0 (min.)	1,5 (min.)	6,0 (max.)	12,0 (max.)

B. Plywood

# (a) Interior-Grade Plywood:

An unfortified tannin-formaldehyde adhesive resin was developed for bonding wood veneers in the manufacture of interior- or semi-exterior-grade plywood. The glue mix was prepared as follows:

#### Glue mix.

Constituents	Parts by Mass
Wattle extract	100
Water	100
Caustic soda	1,15
Coconut shell flour (200 mesh)	10-15
Paraformaldehyde (fine powder)	10

The extract (100 parts) was dissolved in warm water (100 parts) containing caustic soda (1,15 parts) and allowed to stand overnight to ensure full hydration of the material.

To the solution of extract, at pH = 7.0-7.2 was added a hardener component, consisting of a mixture of paraformaldehyde powder (10 parts) and coconut shell flour (10-15 parts), shortly before use.

The effective pot-life of this unfortified plywood resin was approximately 4 hr. at  $20^{\circ}\text{C}_{\bullet}$ 

#### Assembly and pressing conditions.

	• -	
Veneer moisture content Glue spread	Range: Hardwoods:	4-10% 150-200 g./m. <sup>2</sup> s.g.l.* 200-250 g./m. <sup>2</sup> s.g.l.*
Open assembly time	Softwoods: Range:	15-60 min.
Pre-pressing (cold)	Premoure:	9 kg./cm. <sup>2</sup>
	Temperature:	ambient
	Time:	5-10 min.
Hot-pressing	Pressure:	12 kg./cm. <sup>2</sup>
	Temperature:	120°C. (min.)
	Time:	5 min. basic, plus 1 min. per
		mm. to furthest glue line.
Post suring	Hot stack nam	els for 24-48 hr.

Post-curing
\* S.g.l. = single glue line.

After post-curing, the panels were conditioned at 20°C. and 60% Relative Humidity for 24-72 hr. prior to testing. Physical testing was conducted according to B.S. 1455 (1963)(14), using the knife test, after soaking the plywood specimens in cold water (20°C.) for 24 hr. or boiling samples for 3 hr. The knife-tested panels were evaluated by comparison of their bond qualities with the Master Scale (1-10 rating) for assessing plywood.

Average results of Okoumé plywood specimens are summarized in Table 2.

Table 2

Average Bond Qualities of Okoumé Plywood Panels

using the Knife Test

Veneer Species	Knife Test Results		
	Cold Soak (24 hr.)	Boil (3 hr.)	
Okoumé	7 (min. 4)	5 (min. 2)	
Tropical hardwoods B.S. 1455 (1963) - BR Bonding	5 (min. 2)	5 (min. 2)	

# (b) Exterior-Grade (Marine) Plywood:

In order to produce plywood suitable for use in outdoor applications, e.g. marine plywood, it was necessary to fortify the tannin-formaldehyde formulation with a phenol-resorcinol-formaldehyde resin.

Use was made of factorial experiments to develop a suitable adhesive for the production of marine-grade plywood<sup>(7)</sup>. Glue mix variables, assembly and pressing conditions were optimized by means of a series of factorial experiments, coupled with computer-assisted interpretations of shear test and ply adhesion results.

The fortifier was prepared as follows:

# Fortifier.

Constituents		Parts by	Mass
Phenol Formaldehyde Caustic soda Resorcinol Phenol	(40% aqueous) (14% aqueous)	100 86 18 46 60	

Phenol (100 parts) was dissolved in formaldehyde solution (86 parts, 40% aqueous) and caustic soda solution (18 parts, 14% aqueous) was added. The mixture was heated with stirring at 90°C. for 60 min. After cooling the solution to 70°C., resorcinol (46 parts) and phenol (60 parts) were added and the resultant mixture refluxed for a further 60 min. at 90°C., before cooling it to ambient temperature.

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The glue mix was composed of the following:

## Glue mix.

Constituents	Parts by Mass
Wattle extract	100
Water	80-90
Caustic soda	1,5
Fortifier	24
Sodium pentachlorophenate	0,2
Coconut shell flour (200 mesh)	10
Paraformaldehyde (fine powder)	12

The extract (100 parts was dissolved in warm water (80-90 parts) containing caustic soda (1,5 parts) and allowed to hydrate overnight. To this solution was added sodium pentachlorophenate (0,2 parts) as antifungal agent and the fortifier (24 parts). The pH of this resin was in the range 7,5-7,7.

In this form the fortified wattle based resin solution was ready for use with the hardener component. This liquid formulation could be spray dried, however, to yield a powdered resin, having indefinite shelf-life. It was catalysed in the normal way with the hardener, consisting of fine powdered paraformaldehyde (12 parts) and coconut shell flour (10 parts).

The pot-life of this fortified tannin-formaldehyde resin was approximately 4 hr. at 20°C.

Assembly and pressing conditions were similar to those used for the construction of interior-grade plywood. Post-curing and conditioning were done as before and the plywood specimens were knife-tested according to BS. 1455 (1963)<sup>(14)</sup>.

Typical knife test results of plywood panels made with Okoumé, South African Pine or sandwich constructions of these veneers are shown in Table 3.

<u>Table 3</u>

Average Bond Qualities of Marine Plywood Panels

Veneer Species		Knife Test Results		
Back	Core	Face	Cold Soak (24 hr.)	Boil (72 hr.)
Okoumé Okoumé S.A. Pine	Okoumé S.A. Pine S.A. Pine	Okoumé Okoumé S.A. Pine	8 (min. 5) 8 (min. 5) 7 (min. 2)	8 (min. 4) 8 (min. 7) 6 (min. 4)
Tropical har B.S. 1455	ardwoods (1963) - WBP	Bonding	5 (min. 2)	5 (min. 2)

# C. Shutteringboard

Shutteringboard or blockboard consists of a central core of wooden blocks, glued with a pva adhesive, laminated between several layers of wood veneer. The product is used extensively in the building trade for concrete formwork or shuttering.

The glue mix consisted of a wattle based adhesive formulation fortified with a phenol-formaldehyde resin, which was prepared as follows:

# Fortifier.

Constituents	Parts by Mass
Phenol	100
Formaldehyde (40% aqueous)	48
Caustic soda (25% aqueous)	10,8

The phenol (100 parts) was dissolved in the formaldehyde solution (48 parts, 40% aqueous) and the caustic soda solution (10,8 parts, 25% aqueous) was added. The mixture was heated at 90°C. for 90 min. and cooled to ambient temperature. The glue mix was prepared as follows:

# Glue Mix.

Constituents	Parts by Mass
Wattle extract	100
Water	80
Caustic soda	1
Fortifier	20
Sodium pentachlorophenate	0,2
Coconut shell flour (200 mesh)	10
Paraformaldehyde (fine powder)	10

The extract (100 parts) was dissolved in warm water (80 parts) containing caustic soda (1 part) and allowed to hydrate overnight. To the extract solution was added the antifungal agent (0,2 parts) and the fortifying resin (20 parts) and the mixture was well stirred. The pH of the resin was between 7,0 and 7,2.

The liquid resin could be used in this form or spray dried to prolong the shelf-life. In either case, the resin was catalysed shortly before use with the hardener, consisting of a mixture of powdered paraformaldehyde (20 parts) and coconut shell flour (10 parts). The pot-life of the final adhesive resin was approximately 5 hr. at 20°C.

#### Assembly and pressing conditions.

These were similar to those used for plywood except the following:

250-300 g./m. $^2$  single glue line. Glue spread Time: 12 min. for 22 mm. assembly consisting Hot-pressing of 16 mm. core and 4 veneers.

After post-curing and conditioning the boards were subjected to a cyclic soak test before being knife tested according to B.S. 1455 (1963)(14). The soak test consisted of 5 cycles of alternate soaking in cold water (20°C.) for 16 hr. and drying at 60°C. for 8 hr.

Typical knife test results of blockboards, produced from S.A. Pine cores and Okoumé cross-bands and face veneers, are summarized in Table 4. -79-

# Average Bond Qualities of Shutteringboards

Knife Test Results						
Face/Cross-band	Cross-band/Core					
8 (min. 5)	5 (min. 2)					
Percentage Edge Delam	ination: 1,5% (max.)					

#### D. Laminated Timber

A chemically modified tannin based adhesive resin, containing resorcinol, was developed for use in the finger jointing of short lengths of timber and in the lamination of strips of wood to form laminated beams.

Wattle bark extract was reacted with formaldehyde to give a tannin-formaldehyde intermediate which was reacted further with resorcinol under suitable pH conditions to give a tannin-resorcinol-formaldehyde cold-setting resin. This resin was spray dried to yield a powdered B-stage resin with indefinite shelf-life. As catalyst for the resin a mixture of powdered paraformaldehyde and inert catalyst, such as wood flour or coconut shell flour, was used.

The cold-setting adhesive resin was prepared as follows:

Constituents		Parts by Mass
Wattle extract Water Methanol Defoamer	} Solution A	64,2 25,8 30,0 0,3
Formaldehyde (40% aqueous)		9,2
Resorcinol Water	} Solution B	30,0 30,0
Sodium Hydroxide Water	} Solution C	0,6 1,8

Solution A was prepared by heating a mixture of water (25,8 parts) and methanol (30,0 parts) to 50-60°C. and dissolving the extract (64,2 parts) slowly with efficient stirring in the aqueous methanol. To solution A was added defoamer (0,3 parts) and the solution heated to 70°C. with agitation. Formaldehyde solution (9,2 parts) was added rapidly to solution A and the resultant mixture heated at 70°C. for 2 hr.

In the meanwhile solution B was prepared by dissolving resorcinol (30 parts) in water (30 parts) which had been preheated to 80°C.

Solution C was prepared by dissolving sodium hydroxide (0,6 parts) in cold water (1,8 parts).

After 2 hr. at 70°C., solution B was added rapidly to solution A. The heating of the reaction mixture was stopped and solution C was added

over 15-20 min. An exothermic reaction ensued and after this had subsided, heating was resumed. The reaction vessel was converted from a reflux to a distillation set-up and the methanol-water solvent mixture was distilled off until approximately 50 parts had been collected.

Heating was stopped and sufficient water was added to dilute the resin to the required solids content. The pH of the resin was 6,5 and this was adjusted to pH = 7,5-7,9 by addition of 1,5-2,0% of sodium hydroxide solution (25% aqueous), based on the mass of extract solids.

The final resin could be used conveniently in liquid form with the hardener component, consisting of paraformaldehyde (10 parts, on resin solids) and inert filler (10 parts, on resin solids), to produce a coldsetting laminating adhesive. In order to confer an indefinite shelf-life on the resin, it was diluted to 35-40% solids and spray dried. In solid form a separate hardener was provided to give a two-component cold-setting adhesive resin for finger jointing and lamination of timber.

The working life of this adhesive was found to vary with temperature as shown in Table 5.

Table 5

Variation of Pot-life of Cold-setting Adhesive with Temperature

Glue Mix Temperature (°C.)	Pot-life of Glue (hr.)
20	3,0
25	2,5
30	1,5
35	0,75

In practice, glue line temperatures in the range  $20-25^{\circ}\text{C}$ , were found to be satisfactory. Glue spreads of 16-20~kg, per 100~m. of single glue line were used for medium density timber. Closed assembly times in the range 10-40~min, were used, after which the wood strips were clamped at pressures of 10-15~kg./cm. for 14-20~hr, at  $20-25^{\circ}\text{C}$ , to form satisfactory bonds. In order to allow these bonds to develop maximum strength the beams were set aside for a further period of 24~hr, before conditioning and physical testing the samples.

Standard European Beech wood overlap strip tests were conducted with the cold-setting resin according to B.S.  $1204 - 1965^{\left(15\right)}$ . In addition, block shear strength tests were carried out using S.A. Pine and Eucalyptus saligna timber according to S.A.B.S.  $876 - 1967^{\left(13\right)}$ , as the dry shear strength. Finger jointed specimens were tested for failing loads in tension of 25 x 5 mm. specimens, after soaking in cold water for 16 hr. and after boiling in water for 3 hr. Results obtained for these tests are summarized in Tables 6, 7 and 8.

<u>Table 6</u>

Average Bond Qualities and Percentage Wood Failure of Beech Wood

Strips Prepared with Wattle Based Cold-Setting Adhesive

D:	ry	Cold Soak	(16 hr.)	Boil (3 hr.)			
Failing Load N	Wood Failure %	Failing Load N	Wood Failure %	Failing Load N	Wood Failure %		
3460	73	2560	2560 87		100		
B.S. 1204 Requiremen	- 1965(15) nt	2224 (min.)		1446 (min.)			

Table 7

Average Block Shear Strengths and Percentage Wood Failure of S.A. Pine and Eucalyptus Saligna Specimens

Timber	Treatment	Treatment Shear Strength Wood Failure MPa. %				
Pine	Dry Soak Boil	9,9 (4,96 min.) 4,4 3,9	86 90 80	1 (10% max.)		
Saligna	Dry Soak Boil	11.6 (8,27 min.) 6,8 6,2	95 90 90	7 (10% max.)		

Min. and Max. values refer to S.A.B.S.  $876 - 1967^{(13)}$ .

Table 8

Average Bond Qualities and Percentage Wood Failure of Finger

Jointed Specimens of S.A. Pine

Treatment	Failing Load N.	Wood Failure %
Cold Soak (16 hr.) Boil (3 hr.)	3960 (1700 min.)* 4093	74 69

<sup>\*</sup> Min. value required for S.A. Pine.

#### DISCUSSION

#### A. Particleboard

Weather resistant particleboard was manufactured with an unfortified, wattle based tannin-formaldehyde formulation, applied at a level of 10% on the mass of wood chips. The relatively high viscosities of concentrated wattle extract solutions were reduced by alkaline pre-treatment. Addition of 1% by mass of wax emulsion to the wood chips improved the weatherproofing qualities of the board.

During manufacture it was found that wattle based adhesives could tolerate a high level of moisture in the sprayed mat of chips. Final moisture contents in the range 15-20% were commonly used, without attendant eruption or "blowing" of the board on opening the hot press. This feature of tannin-formaldehyde adhesive resins results in a cost saving, because the wood chips need not be dried to as low a level as is common with ureaformaldehyde resins.

In order to obtain a waterproof exterior-grade particleboard it was necessary to increase the amount of adhesive resin to 12-14%, based on the mass of wood chips. This served to improve the internal bond strength and percentage swelling to levels which fully complied with the DIN 68761(1963) Standard (4) for weatherproof particleboard.

Use of a neutral pH level for tannin-formaldehyde particleboard glues renders the product less susceptible to excessive swelling in water than is the case with commercial phenol-formaldehyde resins, which are usually highly alkaline in character.

Tannin-formaldehyde formulations bond rapidly with the wood chips and pressing cycles have been used which are as fast as those used with ureaformaldehyde resins.

#### B. Plywood

Unfortified tannin-formaldehyde resins have been used to produce semi-exterior grade plywood, while phenol-resorcinol-formaldehyde fortifiers have been incorporated in glues for marine plywood. The former adhesive resin gave weather resistant bonds with several veneer species, while the fortified adhesive yielded fully waterproof bonds with hardwoods.

Since wattle tannins have a higher reactivity than phenol it has been possible to increase the production rate of plywood construction, because tannin-formaldehyde resins need shorter assembly and pressing times than most phenol-formaldehyde formulations. Hot post-curing improves the bond strengths of plywood made with wattle based glues.

The neutral character of glue lines of tannin-formaldehyde resins have certain advantages in plywood production. Too high an acidity or alkalinity in the glue line may seriously affect the long term durability of products from wood.

#### C. Shutteringboard

Wattle based resins, fortified with a phenol-formaldehyde formulation, have been used in the production of blockboard or shutteringboard.

The bond qualities of such boards comply with the specification of BS 1455 (1963)(14) for plywood made from tropical hardwoods. In practice, shutteringboard is required to be water resistant since it comes into contact with moisture during concrete formwork. In order to render it more waterproof the outer surfaces of the board are treated with oil or other water-repellent agents.

As in the case of plywood, tannin-formaldehyde formulations show certain advantages in this application. Neutral glue lines and speedier assembly and pressing conditions are the most important of these advantages.

#### D. Laminated Timber

Resorcinol has the unique property, among phenols, of producing a cold-setting adhesive resin with formaldehyde under neutral conditions (6). This fact has been utilized in the development of resorcinol based adhesives for finger jointing of short lengths of timber and for lamination of strips of wood to produce structural beams, at ambient temperatures.

As a result of the gradual increase in cost of resorcinol over the last few years and its recent scarcity due to the petro-chemical crisis, it was found necessary to investigate the use of wattle tannins in cold-setting adhesive formulations in South Africa.

A chemically modified, wattle based, resorcinol-tannin-formaldehyde resin has been developed for use with South African Pine timber. This adhesive resin has passed the B.S. 1204 - 1965 Specification (15) for laminating adhesives on European Beech wood and has received full approval for use with S.A. Pine timber. Block shear tests, according to S.A.B.S. 876 - 1967(13), and tests on finger jointed specimens were conducted on samples made from S.A. Pine and Eucalyptus saligna timber.

In view of the fact that a proportion of resorcinol is used in the preparation of this adhesive, it shows all the characteristics of a purely resorcinol adhesive, but may be produced at substantially reduced cost. This is due to the fact that at present wattle extract is only 10% of the price of resorcinol in South Africa.

The flavanoid tannin molecule is composed of units having resorcinol-type reactivity in the A-rings. This characteristic is utilized in the production of copolymers of tannins and resorcinol with formaldehyde to give fully waterproof cold-setting timber adhesives.

#### CONCLUSIONS

Wattle based tannin-formaldehyde adhesive resins have been used in commercial manufacture of particleboard for several years in South Africa and Australia. Board producers in Australia make use of a resorcinol-formaldehyde fortifier in their particleboard resin<sup>(11)</sup>, whereas the South African formulations are unfortified. Such fully waterproof particleboard has found application in the manufacture of freeway road signs, as exterior cladding for temporary buildings, as floor underlays, as ceiling materials and in the general building trade.

At present the market for plywood in South Africa is relatively small. Interior-grade plywood is used mainly in the manufacture of furniture and for decorative purposes. In this application urea-formaldehyde adhesives are commonly used. For semi-exterior uses, however, the unfortified wattle based formulation has shown certain advantages, because of its weather resistant qualities.

In cases where waterproof plywood is needed, such as marine and exterior-grade plywood, the fortified tannin-formaldehyde adhesive resin compares very favourably with phenol-formaldehyde formulations, with regard to pH characteristics and speed of bonding.

A small tonnage of this exterior-grade plywood adhesive has been sold. It has been evaluated by several independent orginisations who have expressed their full approval of its performance. Long term durability trials and accelerated ageing tests have shown that this resin meets all the requirements of an exterior-grade glue.

The tannin-formaldehyde shutteringboard adhesive is the latest in the series of thermosetting resins developed by the Leather Industries Research Institute. Laboratory evaluations have shown the resin to be well suited to this type of application. Full scale factory trials remain to be completed before this adhesive resin can be adopted by the local blockboard industry.

The development of relatively inexpensive laminating adhesives, which will set at ambient temperatures and at neutral pH, is of significance in times when raw materials derived from petroleum are in short supply.

Wattle based cold-setting formulations have been shown to be suitable for use in the finger jointing and laminating industries in South Africa. A substantial tonnage of the resin has been produced specifically for the manufacture of glulam beams from South African Pine and Eucalyptus timber.

In all the above developments, it has been one of the main aims to produce these adhesive resins in spray dried solid form. As such, a resin has an indefinite shelf-life.

The bark extract of the Black Wattle tree is an abundantly available, inexpensive raw material for the manufacture of a wide range of adhesive resins for products from wood. Adequate future supplies of this commodity have been ensured through continued afforestation of <a href="Acacia mearnsii">Acacia mearnsii</a> by wattle growers under the control of the South African Wattle Bark Industry.

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# PEROXYACID BONDING OF WHITE FIR WOOD (ABIES CONCOLOR)

Conventional bonding systems utilize a film-forming resin, which, with curing, provides the link between surfaces. Oxidative bonding chemically involves the surface of wood, forming the joint with primary covalent bonds as the link between substrates.

Peroxyacetic and peroxypropionic acids were used to laminate sawn white fir veneer. Variables used in this experiment include hydrogen peroxide concentration, time for peroxyacid formation or "pot life" and mineral acid catalyst for peroxyacid formation.

It was generally observed that higher hydrogen peroxide concentrations, longer pot lives, and the more reactive organic acid led to higher shear strengths, when tested both wet and dry. Shear strengths of 828 psi dry and 304 psi wet were recorded, this compared to 1500 psi dry and 600 psi wet for phenolic controls.

In general this experiment developed data exhibiting a wide range of variability. No immediate reason for this variability was obvious, but may involve the lack of gap-filling capability inherent with this type of bonding.

#### ROBERT K. KRUEGER

Bob is now Senior Research Chemist at the Krause Milling Co., Milwaukee, Wis. His current areas of research are concerned with the utilization of corn and sorghum milling products and byproducts in brewing and industrial applications. He received a BS degree in chemistry in 1963 from the Indiana Institute of Technology at Fort Wayne. He has done graduate work in organic and physical chemistry at Florida State University, Tallahassee and Marquette University, Milwaukee. Prior to joining the Krause Milling Co., he was Section Head of Organic Chemistry at the Joseph Schlitz Brewing Co. and Laboratory Manager of Col Tec Division of Nutrico, Inc.

# AMYLACEOUS PHENOL-RESIN EXTENDERS FOR HIGH-DENSITY PARTICLEBOARD

The particleboard industry originated with the development of inexpensive synthetic resins. The cost of particleboard is greatly influenced by the resin cost, which frequently represents the largest portion of raw material costs. With the advent of the Arab oil embargo and the impending world oil shortages, both the cost and availability of synthetic resins, and especially phenol-based resins, have become major problems for the particleboard industry.

In the case of solid phenolic resins such as used in high-density particleboard, resin costs can be reduced and resin supplies can be extended by the use of a gelatinized amylaceous material modified with a nitrogen-containing compound such as urea. We have found that 20% and more of a solid phenolic resin can be replaced by a gelatinized corn flour containing urea, without impairing the physical quality of the finished particleboard.

Physical properties of both experimental and commercial particle-boards produced with these extended resin systems will be compared to properties of boards made with unextended resins.

# DURABILITY

OF

# ADHESIVES

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#### DICK CASTER

Dick is Senior Scientist in Polymers, Resins, and Adhesives Research Department for the Weyerhaeuser Company, Seattle, Washington.

# PRELIMINARY CORRELATION BETWEEN WEATHERING TESTS AND AUTOMATIC BOIL MACHINE RESULTS

A test procedure consisting of 20 boil—dry cycles per day for up to 40 days has recently been incorporated into an ASTM test method. The development and use of this procedure is described. Examples are given showing how adhesives can be ranked, how processes can be optimized, and how product durability can be predicted. An initial explanation is given showing how to correlate the Automatic Boil Test data with exterior exposure data. Some problems with the exterior exposure data being collected are described.

# PRELIMINARY CORRELATION BETWEEN WEATHERING TESTS AND AUTOMATIC BOIL TEST RESULTS

By Dick Caster and Denise Kulenkamp

# INTRODUCTION

Many new adhesive formulations and types are being considered for use by the wood industry. Large numbers of wood adhesives and new uses envisioned require an accelerated method predicting exterior durability of the bond. This method must not only work but be acceptable to federal and local regulatory and code agencies.

To date, no entirely satisfactory method exists for correlating accelerated test data with actual long-term exterior exposure data. The desired technique would enable us to use an accelerated test method to predict actual exterior exposure performance.

Several organizatons are proposing different types of accelerated aging test methods as being the most suitable; some of which you will hear in the following talks. This discussion presents a proposed model for using Automatic Boil Test data to compare with experience and exterior exposure data.

I would now like to discuss the approaches being used as well as some of the problems encountered. These approaches all used the Automatic Boil Test as the accelerated testing method. In most cases I will not go into detail by including the test data.

# AUTOMATIC BOIL TEST

The Automatic Boil Test has recently been approved by ASTM for inclusion in their manual of test methods. It will be known as ASTM D 3434, Standard Recommended Practice for Multiple-Cycle Accelerated Aging Test (Automatic Boil Test) for Exterior Wet-Use Wood Adhesives.

The test specimens are glued with the recommended processing conditions and the wood species desired tested for 40 days in the Automatic Boil Test and then based on performance and correlation equations, a prediction is made on long-term durability. To develop this technique and the required equations, data from the accelerated aging test method must be correlated with exterior exposure data for identical specimens. The relationship between the accelerated aging data and the exterior exposure data can then be developed. When a new adhesive is developed it is only necessary to conduct the accelerated aging portion of this study and then predict the exterior durability.

In the late 1960's, SCATA (the Stearing Committee for Accelerated Testing of Adhesives) evaluated several accelerated multiple-cycle testing methods. This was primarily done with the cooperation of the Canadian

Forest Products Laboratory, Oregon State University, and the Weyerhaeuser Company. The test which later became the Automatic Boil Test most nearly ranked the ten adhesive types evaluated as would be expected for long-term durability. The Automatic Boil Test consists of the following cycles:

Cycle 1: Ten minutes in boiling water. Cycle 2 and beyond:

- a. four minutes exposed to forced-circulation room temperature air.
- b. 57 minutes exposed to forced-circulation, 225°F air,
- c. ten minutes in boiling water.

The above cyclic procedure gives 20 cycles per day. Tests are carried out through 40 days giving a total of 800 cycles. Samples are pulled at periodic intervals so a rate of performance change curve can be drawr. This curve can then be used to compare with other adhesives and for solid wood. The specimens are tested wet.

# Test Specimens

Specimen design plays an important part in testing glue line durability. Ideally, the wood itself should show no signs of strength loss enabling all of the reduction in performance to be attributed to reduction in glue line durability. It is impossible to design a specimen which does not experience loss of wood strength during accelerated testing. There is also some loss of wood performance in actual real world exposure of glue line and wood combinations. This will be discussed later as it turned out to be a problem in some of our exterior performance evaluations.

Work done by Dee Strickler of Washington State University and ourselves has developed what we feel is the best specimen design. Thin specimens of laminations with l"xl" glue lines offer optimum resistance to stressing in the wood. See Figure 1 for the selected specimens. It is felt that this type of specimen using lumber rather than veneer, as would normally be expected with this specimen configuration, gives a specimen which shows the optimum stress changes between boiling and drying and yet produces very little checking in the wood.

# Exterior Exposure Sites

Since the behavior of the glue is somewhat dependent on the climatic conditions at the use locations, the test method must include this variable for exterior exposure. Such things as radiation, heat, cold, moisture from both rain and humidity and microorganisms should be considered. Difficulty occurs in trying to conduct exposure tests for all of the possible combinations. The following four sites were selected as including the most aspects of climate.

	Comment						
Site	Winter	Summer					
Electric Mills, Miss. Modesto, CA Seattle WA Albert Lea, MN	mild, rain warm, dry mild, wet cold, snow	hot, humid hot, dry mild, wet mild, humid					

The test racks used to hold the specimens are shown in Figures 2 and 3.

# Equation Comments

One of the first steps in correlating the data is determining the equation (or equations) that best fit the data. Changes in shear strength for both Automatic Boil Test and exterior exposure are much greater and more statistically significant than those for wood failure. Most of the discussion to follow is aimed at shear strength.

I don't really have time in this talk to discuss the mathematics used in developing the reasons for selecting the final equations other than to say that data from about 25 adhesive systems for 800 Automatic Boil Test cycles and six-year exterior exposure were used. Also, in order to compare the adhesives, it is easier if they all start with the same initial strength. This is done by converting all shear strengths to percent of original shear strength. The hardening point for each shear strength is then 100. All changes for each adhesive can be compared with this one point (see Figure 4).

Normally we have found two equations are required for the Automatic Boil Test data.

- 1.  $y=C+Dx: 0 \ge x \ge 20$ , x is in cycles, y is in percent of original shear strength.
- 2.  $y=Ae^{Bx}, x \ge 20.$

For exterior data one equation can be used for all data, Y=Ae<sup>Bx</sup>.

For simplifying the discussion to follow, I have often used the  $y=Ae^{BX}$  equation for Automatic Boil Test data. When talking in generalities, this is accurate enough but, as I mentioned before, when actually interpreting the data we would use two equations.

# Automatic Boil Test Data

The Automatic Boil Test Data tends to break the adhesive types down as shown in Figure 5. Adhesives shown represent the best results for that particular type. For example, we have listed several epoxies which give a range as shown in Figure 6 from the best to the poorest.

Solid wood itself will show a loss of strength through the 800 cycles. It is currently being assumed that for an adhesive to have full exterior durability with no failure, it should come quite close in performance to the wood curve. Phenol resorcinols are the only ones which approach this. Some others come close and many adhesives which are being used for exterior applications fall in ranges considerably below this line so this assumption that adhesives should be at the level of the wood curve is somewhat arbitrary. But, since we do have adhesives that follow the wood curve, we are saying this is our goal.

Without talking about how this data correlates with exterior exposure, knowledge of adhesives used in the wood industry would tend to show that these ratings are what would basically be expected. We can, therefore, start using some of this data to evaluate processes and products being made today. The 75% of original shear strength solid wood figure after 800 cycles can be used as a quideline to compare some data.

Many products have been made which pass standard production quality control tests but then fail quite rapidly in service. Four of these will be discussed briefly.

- 1. Keruing and meranti veneer are being used to make exterior grades of plywood siding. The examples we tested in use of exterior phenolic. Both materials after production turned out equally satisfactory in vacuum soak and cyclic boil tests. The keruing plywood begins to delaminate after several years of exterior exposure. The meranti does not. We submitted specimens to the Automatic Boil Test and obtained the results shown in Figure 7. As can be seen from the figure, the Keruing failed very rapidly in the Automatic Boil Test while the Meranti performed at an acceptable level.
- 2. A beam problem which involved the use of an improper gluing and process combination passed the quality control requirements in the product standard. The glue in this application was casein. Again in service, the beams begin to delaminate after 3-5 years. Controls were tested against the bad gluelines (Figure 8).
- 3. Another beam problem also involved the use of the wrong process. Both phenol resorcinol and melamine-urea glues were involved. Again the gluelines were tested after production with the required quality control methods. The results were satisfactory. These beams failed almost immediately in service, some during erection. Controls were tested against the poor gluelines with the results shown in Figure 9.
- 4. Recently we have introduced a phenol resorcinol adhesive on a preheat lam-deck line. As part of determining the optimum processing conditions, samples from different trials were tested with the Automatic Boil Test method. Figure 10 shows an example of durability performance with two different preheat temperatures. 300°F appears to provide better durability.

These four examples appear to show that the Automatic Boil Test is quite a good indicator for predicting improper processing or other problems which might not show up in standard quality control procedures.

# Exterior Exposure Data

The exterior exposure portion of this work started in January 1969. The latest data was obtained in January 1975 which means we have data from up to six years. The original adhesive exposure involved only ten adhesive systems and about forty others have been added at periodic intervals since then. This means that less data is available for some adhesives than others.

Having both ABT and exterior data available enables us to start making some basic comparisons.

# Douglas Fir Versus Southern Pine

Using the right adhesives for which we have the most exterior data available and averaging the results, Table 1 was prepared.

TABLE 1

DOUGLAS FIR VERSUS SOUTHERN PINE\*

		terior ix Year			ABT	(800 (	Tota	Total			
		SS	١	√F	S	S	1	VF.	Aver	Average	
Adhesive	DF	SP	DF	SP	DF	SP	DF	SP	DF	SP	
Avg. of 8	50%	41%	75	54	52	50	87	79	66	56	
Solid Wood	105%	70%	100	100	71	72	100	100	94	86	

<sup>\*</sup>All figures % of original value.

The data shows the adhesives are showing greater deterioration on southern pine than Douglas fir. It also provides data for comparing performance levels after six years exterior exposure and 800 ABT cycles.

# Three-ply Versus Two-ply

We also have data which compare the two-ply parallel grain layups with the three-ply cross grain layups for exterior exposure and Automatic Boil Test.

TABLE 2

PARALLEL VERSUS CROSS GRAIN LAYUP (DF)\*

			or Exposi ar Value)		AB	T (800 (	Total			
Adhesive	S 2 ply	SS WF				SS WF 2 ply 3 ply 2 ply 3 ply			Aver 2 ply	age 3 pl
Avg of 8	50	44	83	71	52	49	90	72	69	59

\*All figures % of original value.

This table shows three-ply to be about 15-20% lower in performance level although the same ranking of adhesives will occur.

# Grain Effect

We also made layups with vertical/vertical grain flat/vertical grain and flat/flat combinations. This is summarized in Table 3.

TABLE 3

VERTICAL VERSUS FLAT GRAIN (DF)\*

	Exterior Exposure (6 Year Value)						ABT (800 Cycle Value)						Total		
	SS WF						SS WF				Average				
Adhesive	VV	VF	FF	VV	VF	FF	VV	<u>VF</u>	FF	VV	VF	FF	$\overline{VV}$	<u>VF</u>	FF
Avg of 8	49	48	46	70	74	80	51	52	50	82	84	76	63	65	63

<sup>\*</sup>All figures % of original value.

This data shows that an almost identical rate of change occurs for vertical, flat or vertical-flat grain layups, even though absolute values may show different results.

# General

The main problem with exterior data after six years is that most adhesives seem to have failed more than I had expected (Figure 4). Solid wood is showing very little change but all glue lines, including those with the phenol resorcinol adhesives, are only at about 60% of original strength. The neat, clean patterns obtained with Automatic Boil Testing also are not apparent. I am not sure if better patterns will become apparent after more years testing.

The goal is to end with a graph showing the Automatic Boil Test data compared to the exterior exposure data, Figure 12. Hopefully one line will be suitable for any and all adhesives.

We have two types of plotting techniques that can be used. The method I call type 1 selects a percent of the original shear strength values, say 90%, and plots that corresponding point as shown in Figure 13. A family of curves is indicated, most follow the line somewhat up the center.

This type 1 method can be repeated for several levels for a percent of original shear strength. The ones being used are 90, 80, 70, 60, and 50 percent. Plotting this data leads to a graph as shown in Figure 14. These graphs appear to be straight lines. This type of technique is known as type 2.

Using the four site average data (only three sites go back six years), a series of curves such as shown in Figure 15 can be developed. The majority of the curves seem to fall into an area roughly in the center. Some adhesives fall in lines radically to either side; i.e., the adhesive looks good in the Automatic Boil Test but is failing under exterior exposure, others looked good for exterior exposure but failed early in the Automatic Boil Test.

This slope is, of course, the key to the whole method. If the adhesives do have the same slope, the Automatic Boil Test time to 70% will give a direct comparison.

At present, the data has not been analyzed enough to understand the exact significance of this correlation slope. The method of placing the adhesive into one of the slope categories from Automatic Boil Test data alone is not yet available.

# Summary

In conclusion, let me say that a detailed test program is underway, only a small portion having been discussed here. The test specimen we have adopted seem to be satisfactory for the Automatic Boil Test but some question has arisen concerning exterior exposure.

The different exposure sites give us different performances which we will be able to attribute to climatic variables. This has not been discussed in this presentation.

The Automatic Boil Test appears to be meaningful and is a severe method for gathering data in an accelerated manner. The Automatic Boil Test also shows much promise as a durability predictor when used by itself as was shown with the process and product evaluations.

Equations are available to handle the gathered data through computer programs. The value of having the correct equations becomes even more apparent, however, when the exterior exposure data has to be extrapolated beyond the present. This again, was not gone into in detail during this presentation but we have attempted to extrapolate the six-year data out through twenty years.

Several miscellaneous items have also come out of this work. These include such things as testing solid wood durability so that the effect of the wood can be subtracted from the wood plus glue combination. To date, we have tried this in only a simple, theoretical manner, but the idea does seem to hold promise. As discussed before, putting the solid wood line on Automatic Boil Test curves and exterior exposure curves also seems to be a good guideline to what is durable and what is not.

The work done to date on correlating the Automatic Boil Test and exterior data has turned out surprisingly well considering the exterior data is only available for six years. If we can figure out what is causing the wide spread on the exterior exposure data and the somewhat inconsistency in rating, we have a meaningful technique. It appears we have the data to do this, it is just a matter of figuring out how to put it together.

While there are still many unknowns, the Automatic Boil Test has proven to be a very useful long-term durability prediction technique. It is possible to develop a technique for predicting durability and I think we will soon have enough test data to accomplish it.

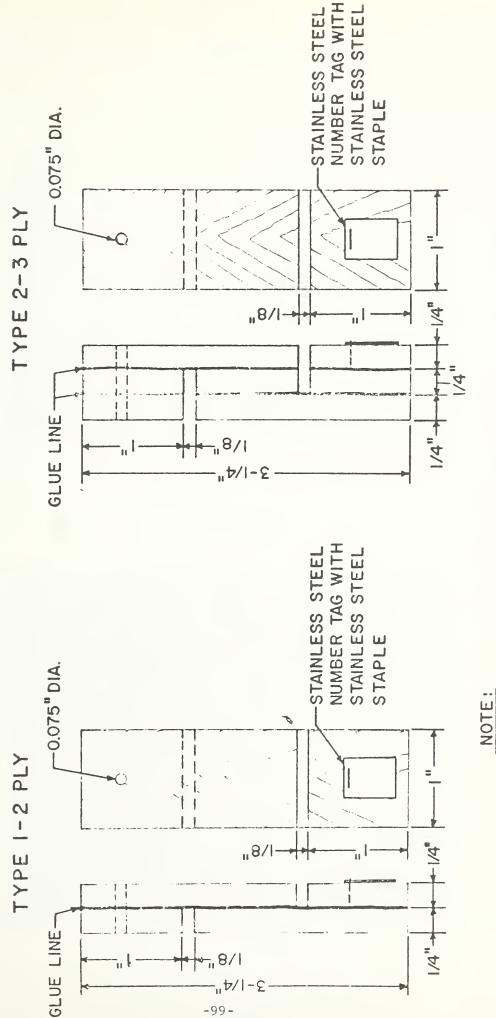
I am open to any comments anyone might have, particularly where you think I am going in the wrong direction. It would also be interesting discussing doing similar work and how we might combine our efforts to exchange data and ideas.

Attach: Figures 1-15

# TEST SPECIMENS

PARALLEL GRAIN PLIES

CROSS GRAIN PLIES



ALL LAMINATIONS MADE WITH 1/4" LUMBER



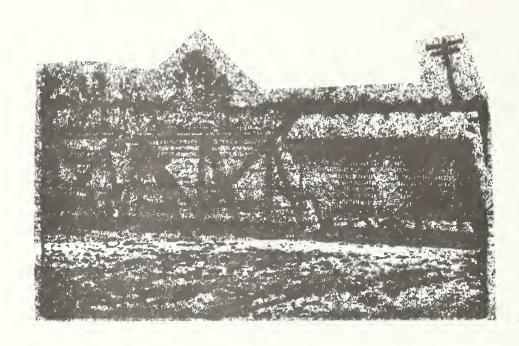
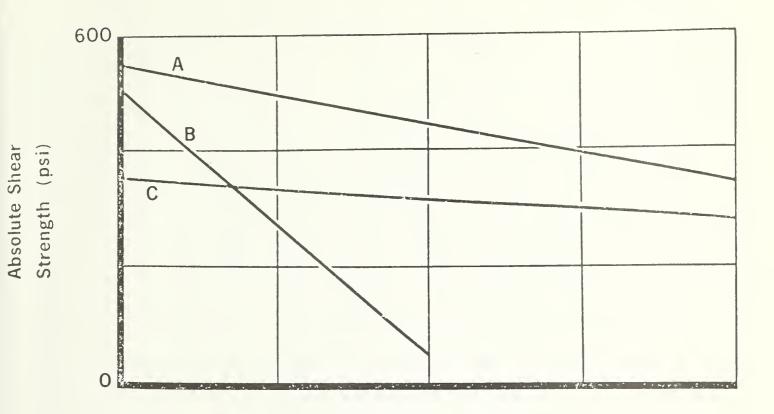
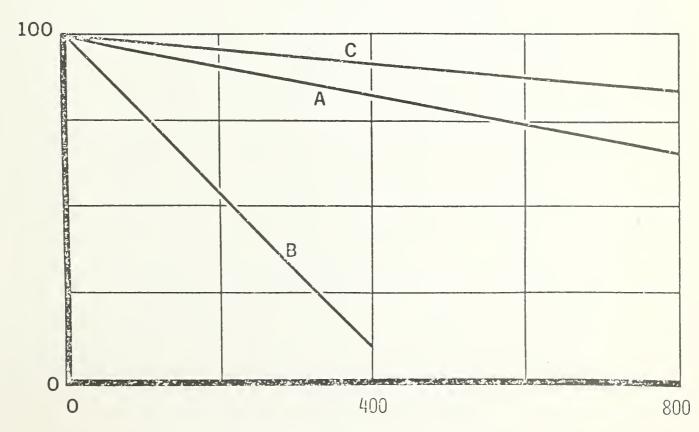




FIGURE 4

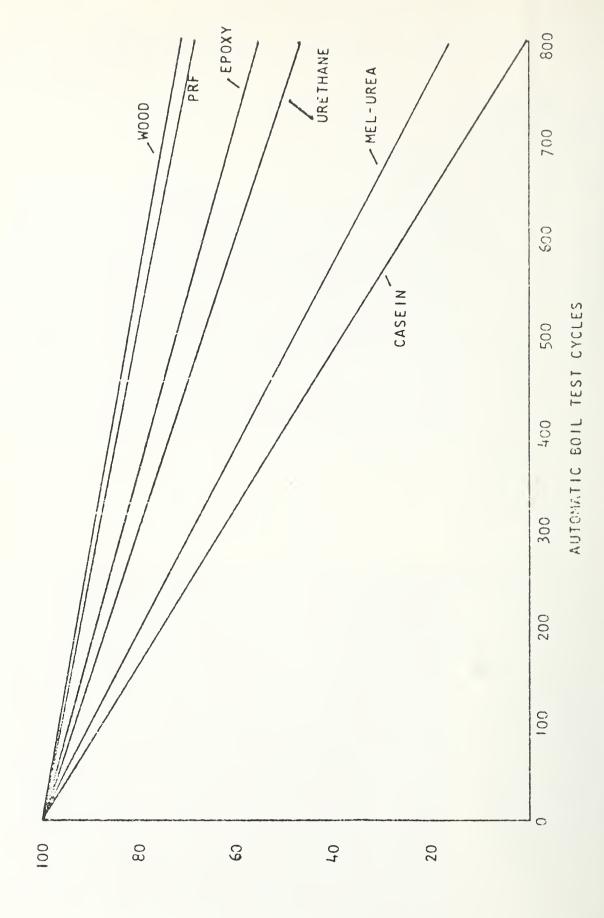
# Use of Percent Original Shear Strength





Percent of Original

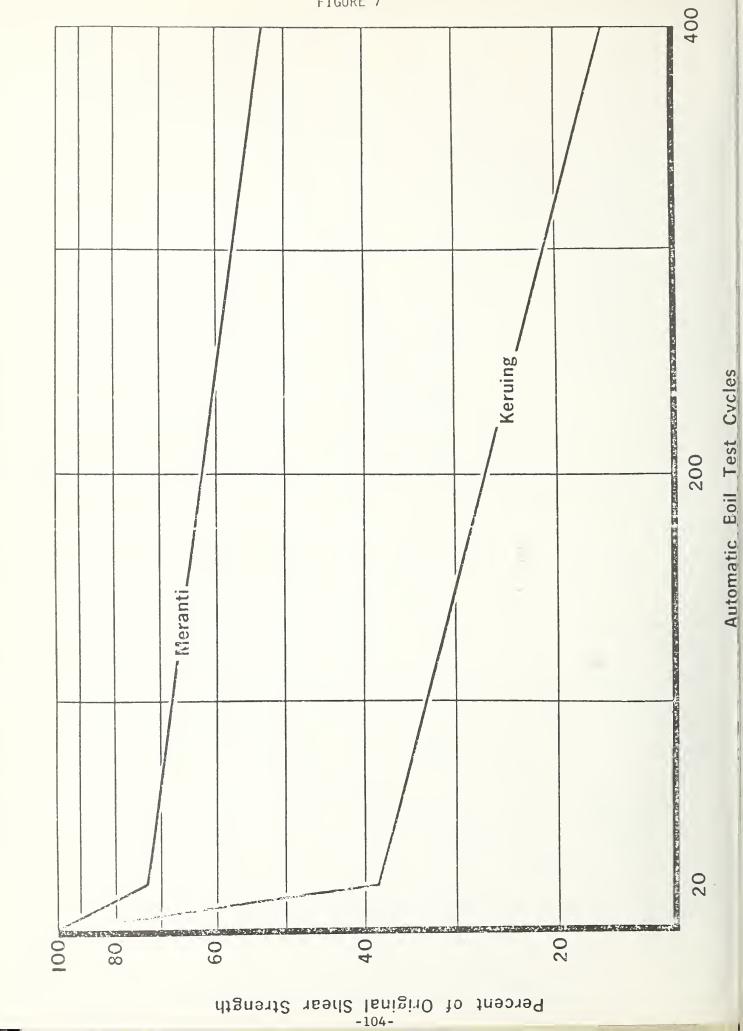
Automatic Boil Test Cycles

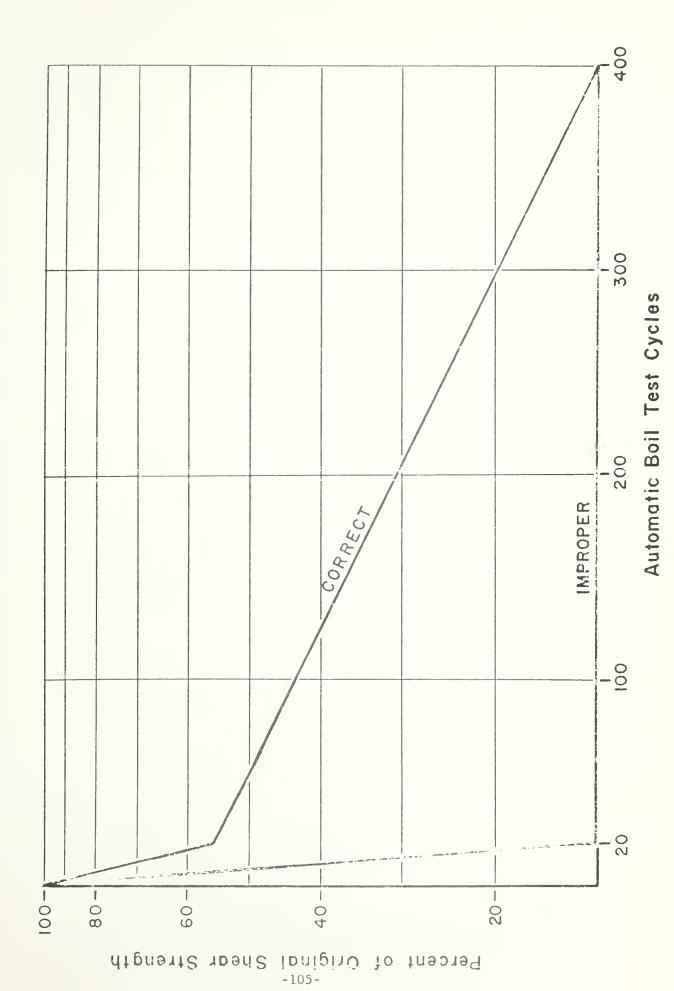


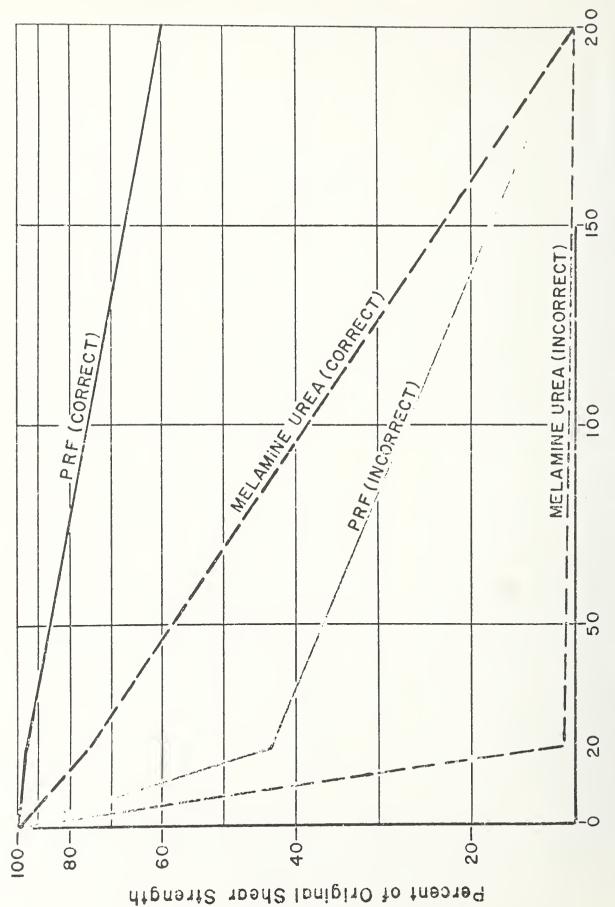
800 -Best Epoxy AUTOMATIC BOIL TEST CYCLES 100 Poorest Epoxy. 100 50 0

FIGURE 6

PERCENT OF ORIGINAL SHEAR STRENGTH







Automatic Boil Test Cycles

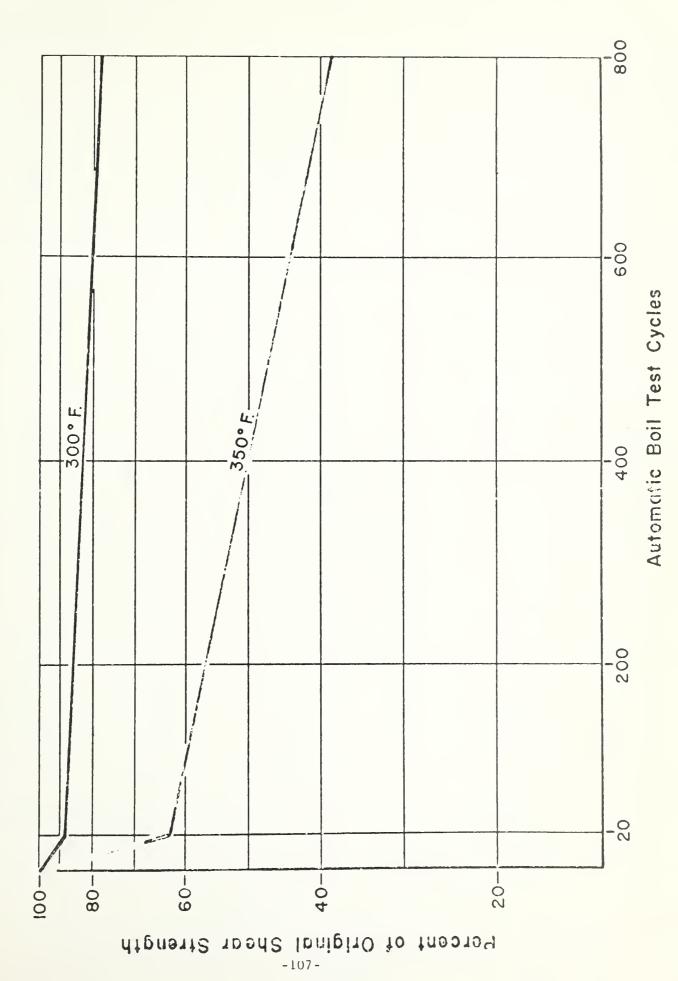
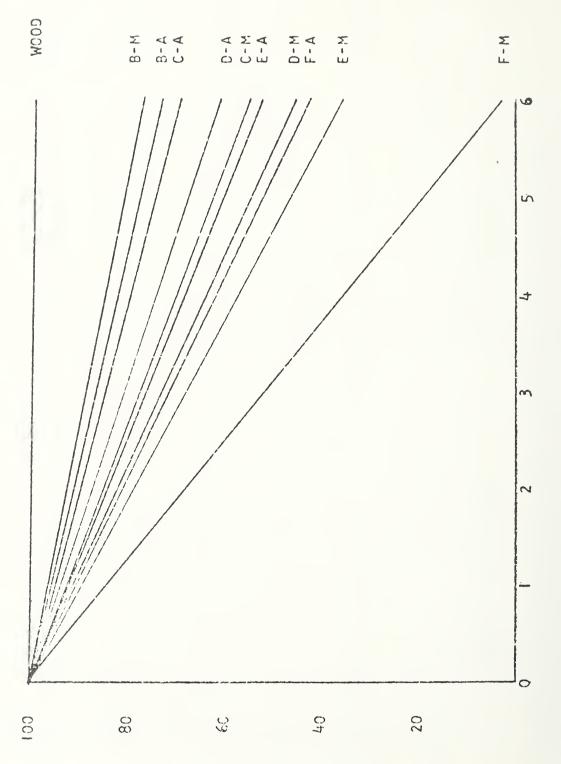


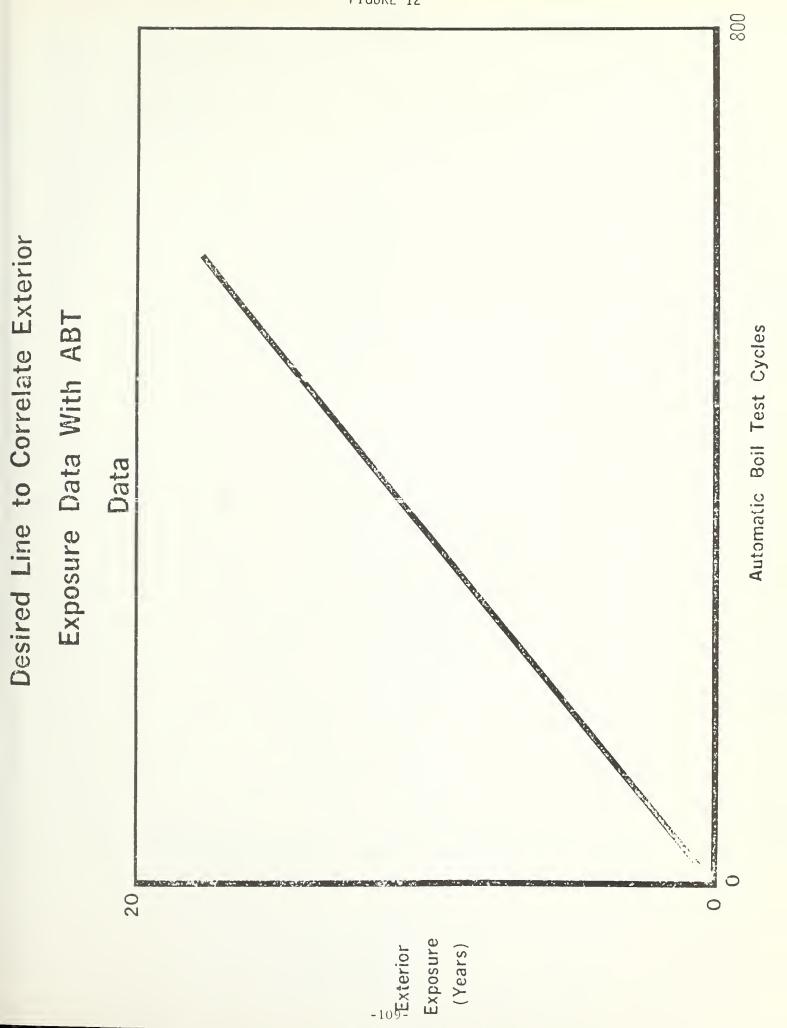
FIGURE 11
SHEAR STRENGTH PERFORMANCE OF VARIOUS
ADHESIVES AFTER EXTERIOR EXPORSURE

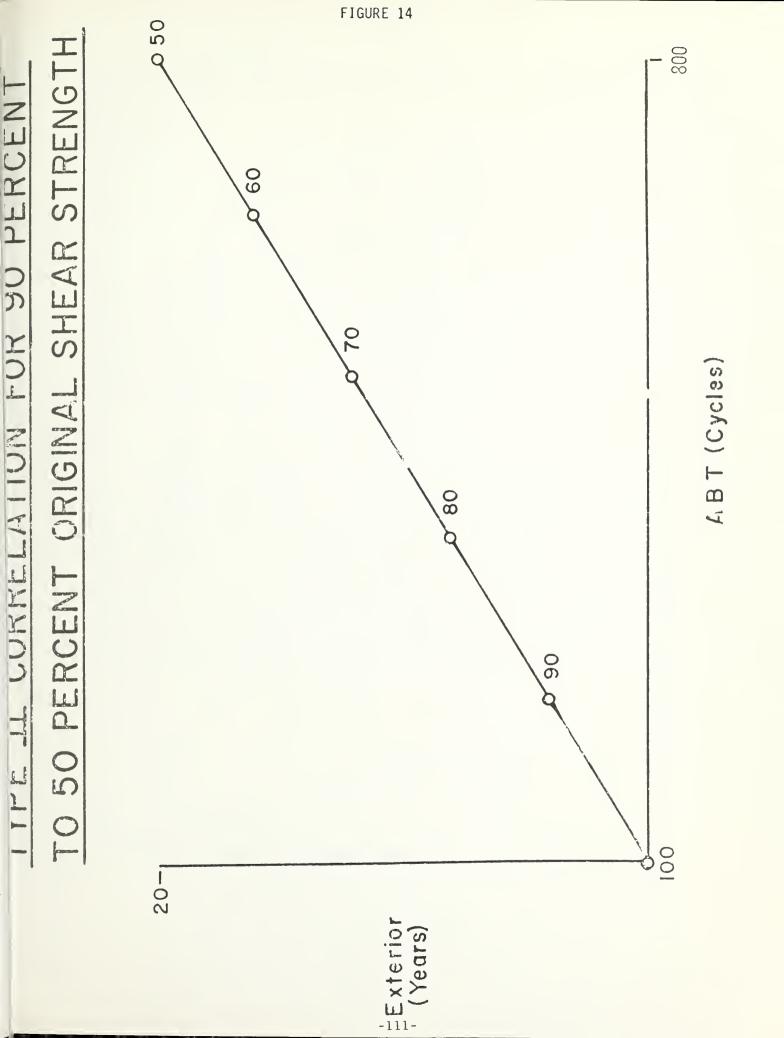


YEASS

STRENGTH

PERCENT OF ORIGINAL





Exterior

Automatic Boil Test Cycles

#### MERRILL A. MILLETT

Mike is a veteran researcher at the Forest Products Laboratory, now working in the area of adhesive systems and adhesive-bonded products. He is one of the real oldtimers at FPL, starting under the C.C.C. program as a student assistant in the summer of 1936 and as a full-time employee in 1941. Mike received his doctorate in physical chemistry at the University of Wisconsin in 1943 and has continued in chemistry-related problems ever since. His major research areas have included dimensional stabilization, wood saccharification, nitrocellulose, analytical development, wood and pulping residues in animal feeds, and currently, the kinetics of wood and adhesive deterioration.

## PRECISION OF THE RATE-PROCESS METHOD FOR PREDICTING LIFE EXPECTANCY

Block-shear specimens of solid maple, phenolic-bonded maple, and melamine-bonded maple were acceleratively aged at six elevated temperatures under both wet and dry conditions and for time periods sufficient to reduce shear strengths to at least one-half of their initial values. Weight-loss and wood-failure data were also obtained. Both loss of strength and loss of weight were found to decrease in an orderly fashion over the entire aging regime and to yield linear Arrhenius relationships for the influence of temperature on the rate of property loss. Graphical or mathematical extrapolation of these relationships provide the key to durability forecasting by the rate-process method. Statistical treatment of the data obtained in the present study was used to provide the upper and lower 95% confidence limits for such durability forecasts.

# PRECISION OF THE RATE-PROCESS METHOD FOR PREDICTING BOND-LINE DURABILITY

By

M. A. Millett, R. H. Gillespie, J. H. Haskell

### Introduction

The efficient use of wood in the form of plywood, particleboard, laminated timbers, and composite structural panels is totally dependent on high-strength durable adhesives. This has created a critical need for short-term accelerated test methods that can provide a quantitative forecast of the long-term performance capabilities of these systems, as well as define their initial acceptability as quality products.

An almost limitless combination of moisture and temperature regimes, to be applied either singly or cyclically, have been suggested or employed for the evaluation of adhesive bonds. A number of these have attained the status of ASTM standards. While the various procedures have proven highly useful for comparison purposes and for in-plant quality control, none of them offers much in the way of predictive capability, i.e., years of expected service based on test performance. This is the hopeful promise of the rate-process techniques now receiving increasing research effort.

The rate-process method has its origin in chemical kinetics, a branch of chemistry concerned with the rates of chemical reactions and with the influence of such variables as temperature, concentration, moisture, catalysts, and the like, on these rates.

Admittedly, the deterioration of cellulose, wood, and wood-adhesive composites is a far cry from the simple reactions normally encountered in textbook illustrations of chemical kinetics principles. Experience has shown, however, that the overall deterioration conduct of these materials does lend itself to interpretation by the basic kinetics techniques. if one follows loss of weight, viscosity, or strength as function of time at a given elevated temperature, the data generally follow an orderly progression, which can be expressed mathematically or at least graphically. By conducting the same experiment at a number of elevated temperatures, one obtains a measure of the influence of temperature on the rate of property loss. From this rate-temperature relationship (generally graphed as an Arrhenius plot) comes the ability to estimate or forecast rates of deterioration beyond experimental feasibility--which is the real essence of the rate-process method.

As we all know, however, extensive extrapolation of experimental data can often lead to grossly misleading conclusions. This was brought home to

us in dramatic fashion in a study of the thermal deterioration of an a acoustic ceiling tile. In this study, we had determined the rate-of-loss of molecules of rupture in bending at three elevated temperatures--130°, 150°, and 170° C. half-life values, computed from the rate curves and plotted as a function of temperature, produced a nice straight-line Arrhenius relationship which, on extrapolation to room temperature, yielded a predicted service life to half-strength of about 10,000 years. Statistical treatment of the data indicated, however, that the upper and lower 95 percent confidence limits of the predicted half-life value could actually range from a negative 10,000 years to a positive 10 billion years--rather distressing spread of values for a forecast of product durability. However, the statistics also showed that if we had used four deterioration temperatures within the same 130°-170° C. range, we could have stated with 95 percent confidence the tile would have an average minimum half-life of 30 years at 30° C. With six temperatures, this minimum half-life projection would have been 200 years.

Thus, with adequate attention to experiment design and control of variables, the rate-process method can be a powerful tool for durability forecasting. The basic objectives of our work, which is part of a cooperative program with HUD, is to determine the influence of experimental variability on the precision of service-life predictions via the rate process procedure and to develop guidelines for attaining maximum precision consistent with affordable experimental time limitations.

## Experimental

Perhaps the best way to approach the experimental details is with an outline as shown in figure 1.

Panel preparation. --In detail, 7- by 20-inch sections were cut from selected sugar maple boards, conditioned to equilibrium at 30 percent RH, and then jointed and planed to thickness--5/8 inch for solid wood and 5/16 inch for bonded wood. The 5/16-inch panels were randomly assembled into two-ply units, spread on the jointed faces with either phenolic or melamine adhesive, and hot-pressed 30 minutes at 200 pounds per square inch at the recommended temperatures. The pressed panels were again brought to equilibrium at 30 percent RH. Sufficient panels were prepared to provide about 1,500 specimens of each type.

Specimen preparation.--By means of a templet, specimen locations were numbered consecutively from 1-1,200 on the panels of each type. As each shear specimen of a given type was cut, it was randomly placed in one of 120 bags. Specimen preparation was completed with the drilling of a 5/16-inch hole in one lip. The ten specimens in each bag were then assembled on tagged wire loops, given a final conditioning at 30 percent RH, and weighed.

Shear specimens were prepared as proposed by Strickler for accelerated aging studies. Dimensio • 1 inch wide by 1-3/4 inches long with 3/8-inch

lips, thus yielding a shear area of 1 square inch. Specimens were compression loaded as in ASTM D 905.

## Accelerated Aging

Dry.--Specimens were mounted on metal racks--7 per aging temperature-each containing 10 wood, 10 melamine, and 10 phenolic specimens for a given time period--and the racks then inserted into preheated chambers capable of  $+0.1^{\circ}$  C. control.

Racks were removed periodically and the specimens placed immediately in desiccators to cool and then weighed. Following a 30-day equilibration at 80° F. and 30 percent RH, specimens were measured and shear strengths determined on a U.T.M. Weight loss data were used to monitor the times for specimen removal during the course of accelerated aging—approximately 12-13 percent weight loss corresponding to a 50 percent loss in shear strength.

Wet.--For accelerated aging in water, all specimens were assembled in groups of 10 on stainless steel wire loops bearing appropriate tags and given a preliminary vacuum-pressure-soak to assure complete water saturation.

Specimen groups were then hung in constant-level water baths controlled to

+0.5° C. Distilled water was used throughout. Loops were removed periodically, the specimens cooled to room temperature measured, and then tested for residual shear strength. Following this, the specimens were again paralleled a 50 percent loss in shear strength for the wood and phenolic-bonded specimens at all temperatures. For the melamine-bonded specimens, a 50 percent strength loss occurred with less than a 2 percent loss in weight.

<u>Data processing.</u>—-Under dry aging conditions, the rate data were best fitted with an equation of the type:

$$\ln S = ae^{bt} + c$$

Where  $\underline{S}$  is shear strength,

 $\underline{a}$ ,  $\underline{b}$ ,  $\underline{c}$  are parameters which describe the model at each temperature, and

 $\underline{t}$  is the time of aging.

The same model fit the wet-aging data for wood and phenolic-bonded wood, but the melamine data were best fitted by a simple straight-line model.

Times to half-strength were computed for each aging condition, using the controls for zero-time strengths, and Arrhenius plots of log half-life versus  $\ell/t$  (absolute) were then regressed and extrapolated to room temperature along with upper and lower 95 percent confidence bonds.

#### Results

<u>Dry aging.</u>—The series of graphs illustrate the rate curves for the aging of solid maple at room temperatures of 120° to 170° C. under dry conditions.

Figure 2 shows shear strength in pounds per square inch at 120° C. versus days of aging. Data points indicate the 10 replicate specimens tested for each time period. Wide scatter typical to all strength measurements on wood. Curve represents the best fit using the model equation shown in an earlier graph. Last data points represent 420 days of aging.

Figure 3, 130° C. Maximum time, 210 days.

<u>Figure 4</u>, 145° C. Includes data for a duplicate run. Maximum time, 110 days.

Figure 5, 160° C. Maximum time, 25 days.

Figure 6, 170° C. Maximum time, 12 days. To show that there is nothing unusual about the aging curves for solid maple, figure 7 is the equivalent 145° C. rate curve for phenolic-bonded maple, and figure 3 the 145° C. curve for melamine-bonded maple.

Figure 9 shows the complete family of curves for the aging of phenolic-bonded maple.

A table of computed half-lives for the three specimen types is shown in <u>figure 10</u>. Reasonable agreement between all three specimen types at any given aging temperature. On the average, each 10° C. decrease in exposure temperature resulted in an approximate 1.8-fold increase in the time to attain half-strength in shear. Half-strength with these maple specimens is still about 1,900 pounds per square inch.

Figure 11, presents a typical Arrhenius plot of the rate data--center curve is the regression line extrapolated to 20° C.--outer curves are the upper and lower 95 percent confidence bonds.

Figure 12, gives the Arrhenius projections for half-life at  $20^{\circ}$  C. along with 95 percent confidence limits at  $20^{\circ}$  C.

Durability of all three specimen types offers no problem under dry aging conditions—Arrhenius projections range from 120,000 years to over 1 million years—even with the wide scatter of the individual data, we can be 95 percent confident that the minimum half—life for shear strength will be at least 8,000 years—a value well in line with the known durability of wood under dry, normal temperature conditions. Equally good durability was provided by the phenolic and melamine adhesives.

Wet aging. -- The next several graphs are representative of the strengthloss versus time curves for aging in water. Figure 13, 100° C., solid maple. Again a wide scatter in the individual data points. Control strengths at zero time average about 1,600 pounds per square inch for all three specimen types.

Figure 14, 85° C., phenolic-bonded, duplicate run.

Figure 15, 60° C., melamine-bonded, duplicate run.

Figure 16 presents the computed half-lives for the three specimen types. As expected, the rates of deterioration of solid maple and phenolic-bonded maple were quite comparable at all aging temperatures—with melamine degrading at 60° C., for example.

Figure 17 shows the Arrhenius projections for half-life at 20° C. along with 95 percent confidence limits.

As with dry aging, we can again appreciate the marked influence of experimental variability on extrapolated estimates of product durability-even though the extrapolations in this case extended only over a modest  $40^{\circ}$  C. temperature range ( $60^{\circ}$  C.  $\rightarrow$   $20^{\circ}$  C.) for the wood and phenolicbonded wood, and a  $20^{\circ}$  C. range ( $40^{\circ}$  C.  $\rightarrow$   $20^{\circ}$  C.) for the melamine-bonded specimens.

With maple, we have a half-life eccese of 400 years at 20° C., but a lower 95 percent confidence limit of 50 years. With phenolic-bonded

maple, the half-life estimate is 1,100 years, but because of greater data variability, the lower confidence limit is 20 years.

With melamine as the bonding adhesive, the 20° C. Arrhenius projection is 6 years, with a 2-year minimum 95 percent confidence limit.

Numerically, the half-strength shear values averaged between 750 to 850 pounds per square inch for all three specimen types.

Using the lower 95 percent confidence limits as a guide, shear strengths of about 800 pounds per square inch would be maintained for at least 50 years by solid maple, 20 years by phenolic-bonded maple, and 2 years by melamine-bonded maple.

#### Summary

In summary, we have taken a comprehensive look at the rate process technique as an experimental tool for forecasting bondline durability. Using some 3,000 block-shear specimens, we have explored the influence of such experimental variables as time, temperature, adhesive type, specimen heterogeneity, and the like on durability forecasts under two environmental extremes—dry and water—soaked.

Statistical treatment of the data indicated a broad error band about the Arrhenius projections. However, use of the lower 95 percent confidence limits provided a positive means for estimating minimum bondline qualities.

Thus, under dry aging conditions, both phenolic and melamine-bonded specimens were essentially equivalent to solid maple in projected durability at 20° C. and could be expected to maintain shear strengths in excess of 1,800 pounds per square inch for at least 8,000 years. Under wet-aging conditions, there was 95 percent assurance of retaining an approximate 800 pounds per square inch shear strength for at least 50 years for wood, 20 years for phenolic-bonded wood, and 2 years for melamine-bonded wood.

Further work remains to be done with statistical treatment of the weight-loss data and interrelating it to the loss of shear strength as well as to the observed extent of wood or glue failure.

Additional information relative to the practical utility of the rate-process method will be obtained from a companion study now under way on the wet and dry aging of solid and melamine and melamine-ureabonded white pine.

## OUTLINE OF EXPERIMENTAL PROGRAM

## Specimen Preparation

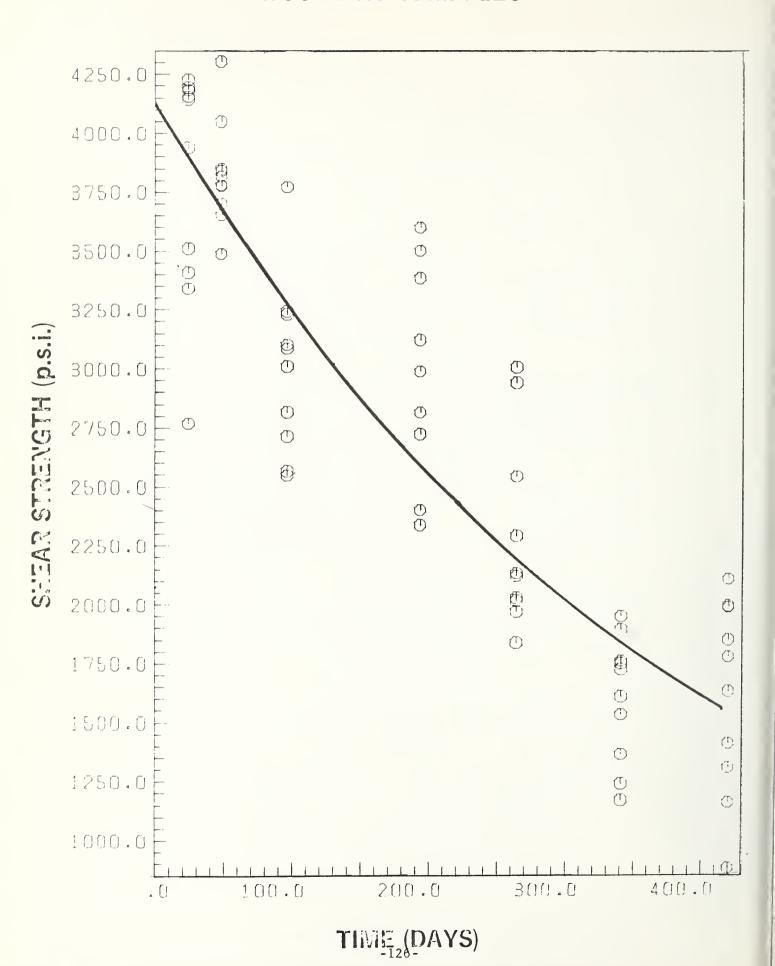
1	Wood	Sugar maple, selected as in
3	Specimen types	ASTM D-905. Solid wood, phenolic-bonded,
1	Test method	melamine bonded. Modified block shear, tested
		as in ASTM D-905.

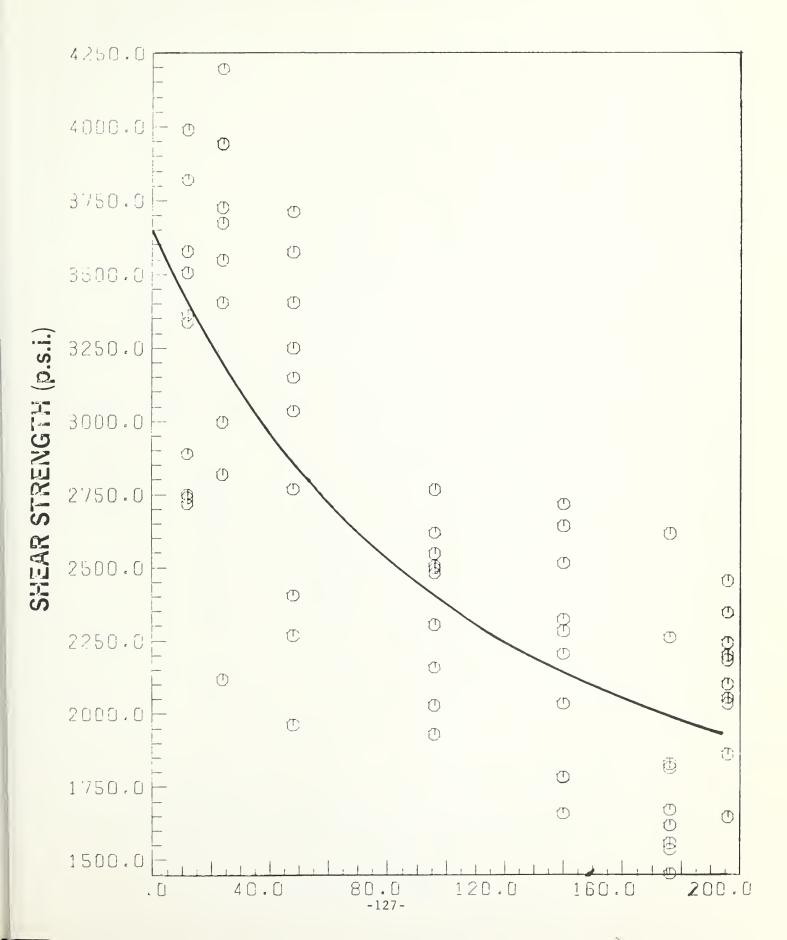
## Accelerated Aging

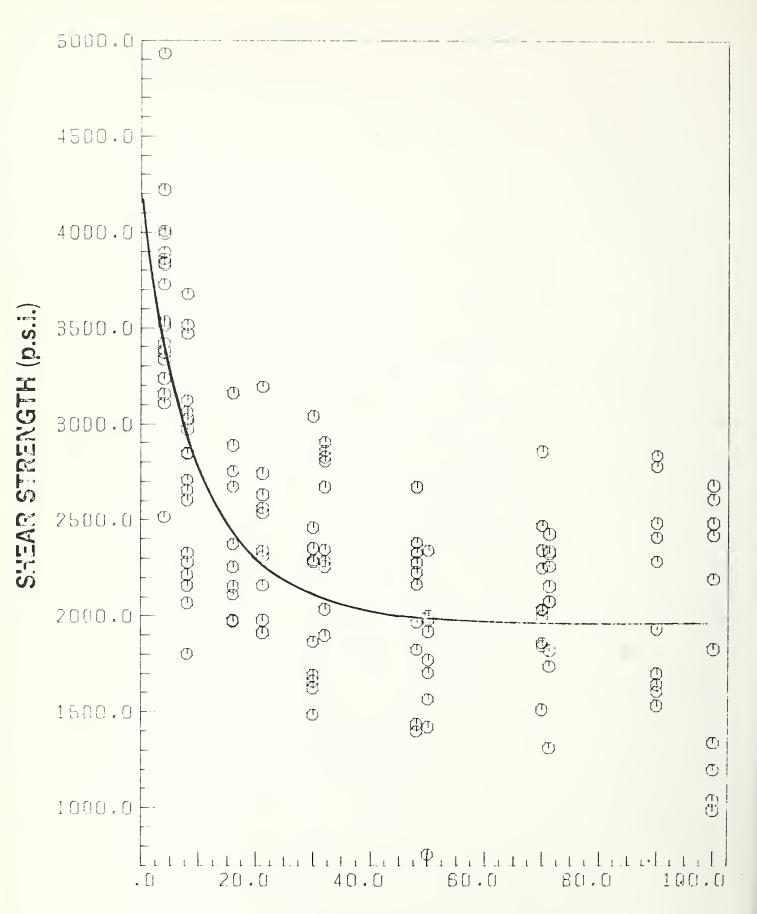
	Aging conditions Aging temperatures	Dry and water soaked.
Ŭ		120°, 130°, 145°-1, 145°-2,
		160°, 170° C.
		60°, 70°, 77.5°, 85°-1, 85°-2, 100° C.
	Melamine	40°, 50°, 60°-1, 60°-2, 67.5°, 75° C.
6	Aging times	To extend beyond half-strength
		in shear.
10	Replications	For each data point.

## Specimen Numbers

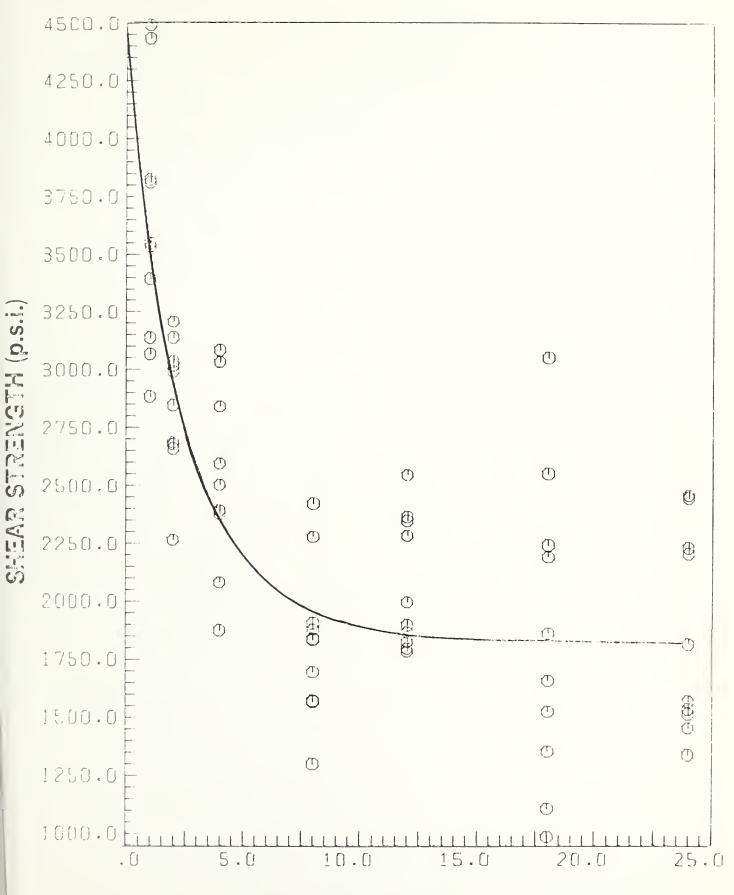
3 Specimen types x 2 aging conditions x 6 aging temperatures x 6 aging times x 10 replications = 2,160 total shear speciments



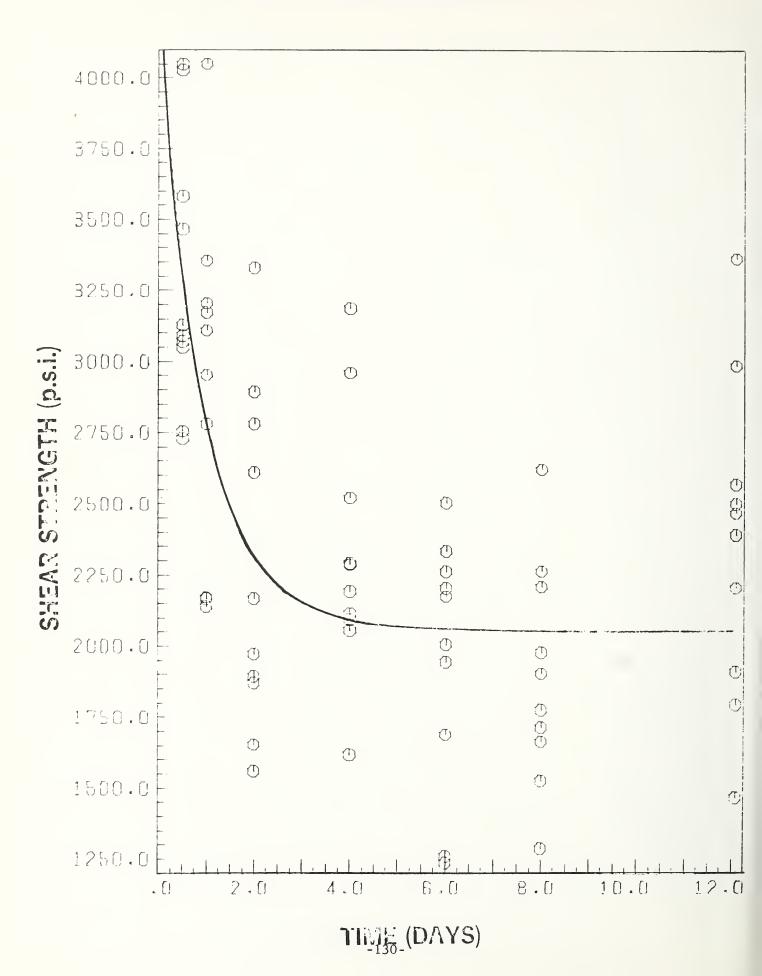




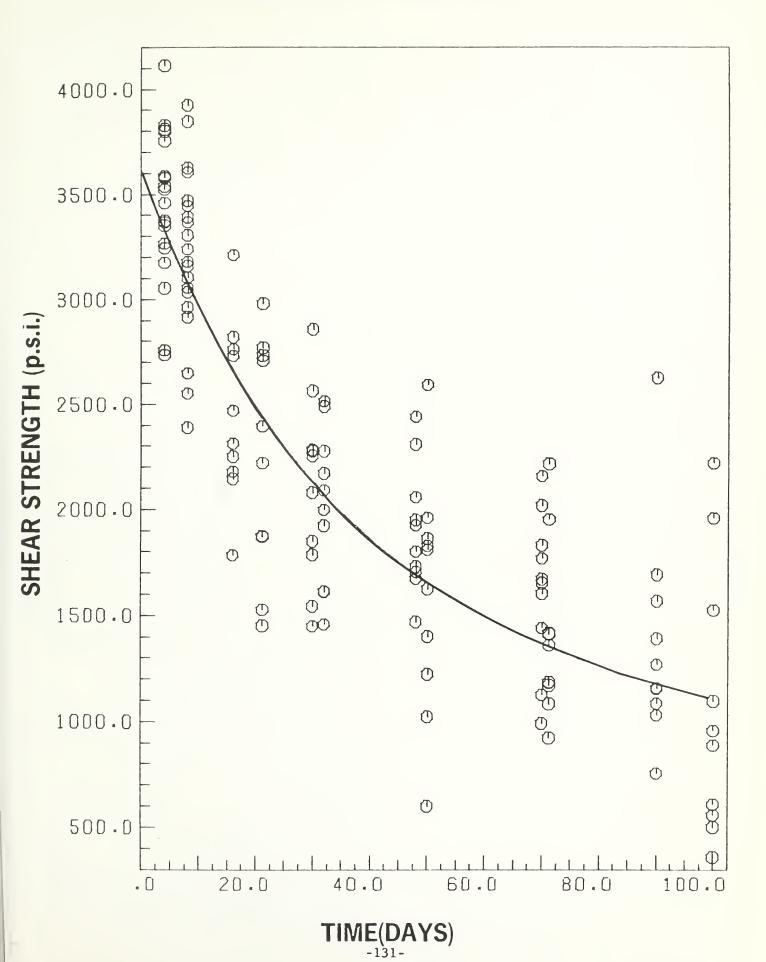
TIME (DAYS)



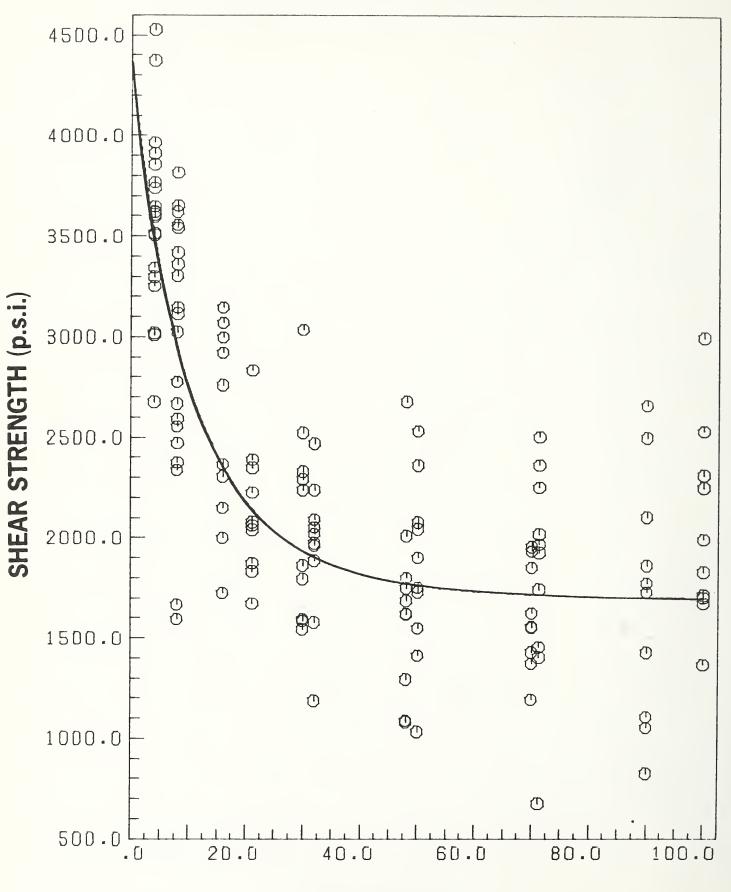
TIME (DAYS)



# PHENOLIC-DRY TEMP. 145

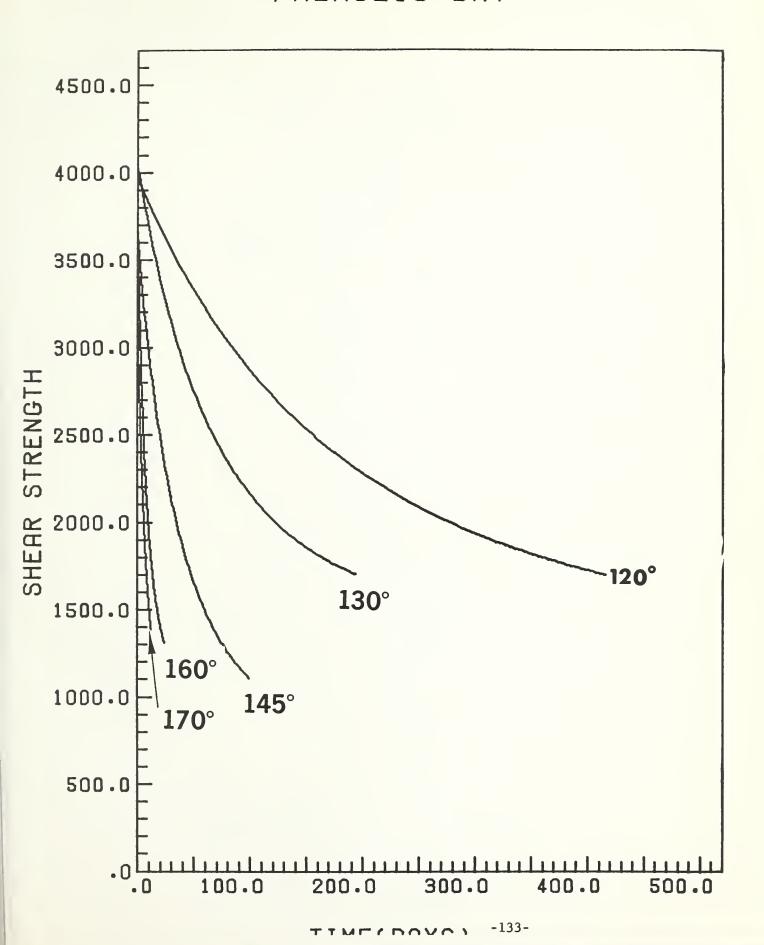


# **MELAMINE-DRY TEMP. 145**



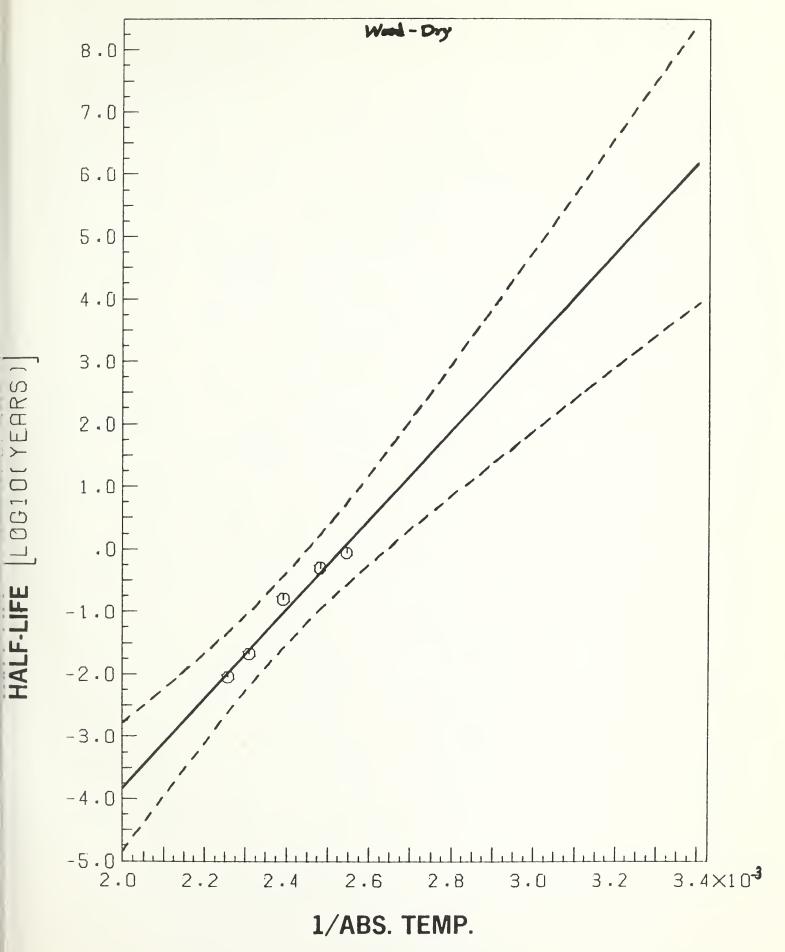
TIME\_(DAYS)

# PHENOLIC-DRY



# OF SHEAR STRENGTH--DRY AGING

Aging Temperature	Tim	rength	
	Wood	Phenolic bonded	Melamine bonded
°C.			
120	310	289	257
130	180	128	158
145	57	36	38
160	8	10	10
170	3	6	6

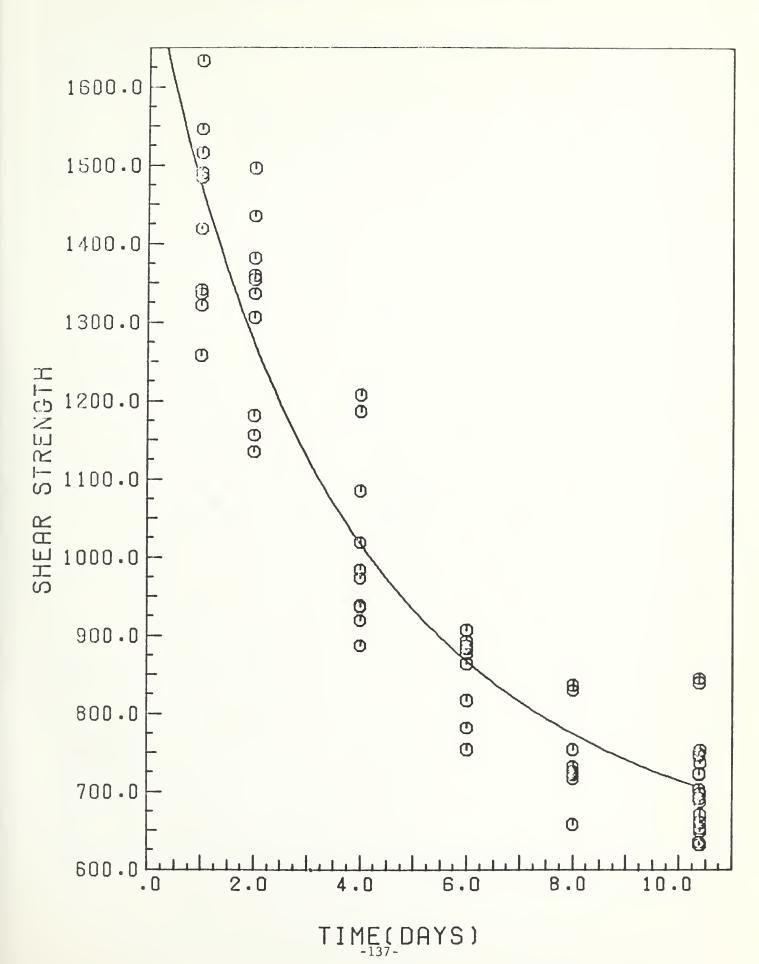


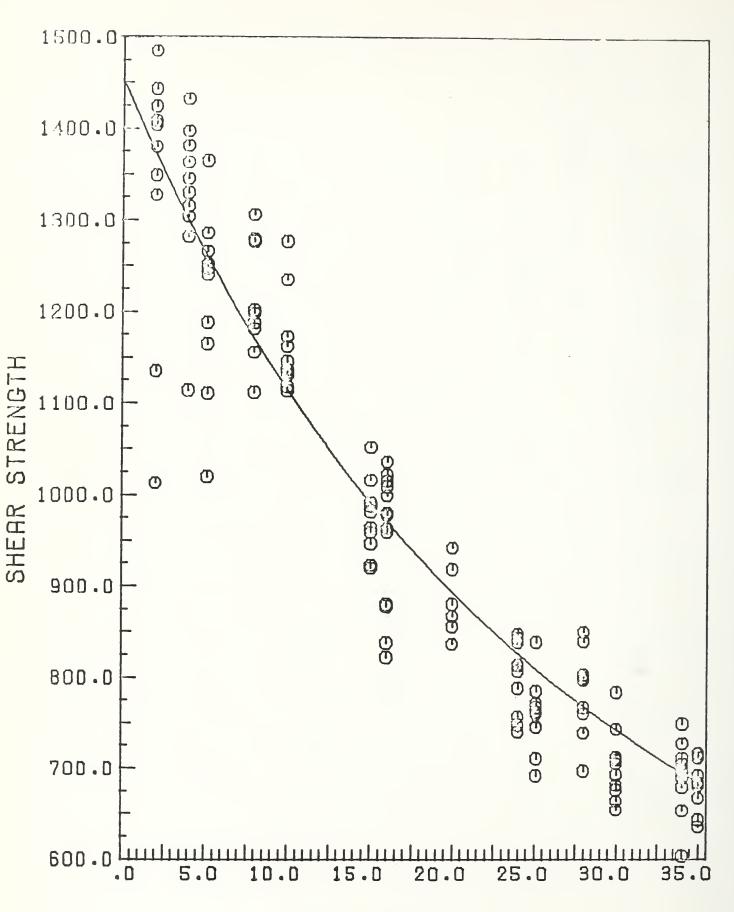
-135-

ARRHENIUS PROJECTIONS FOR

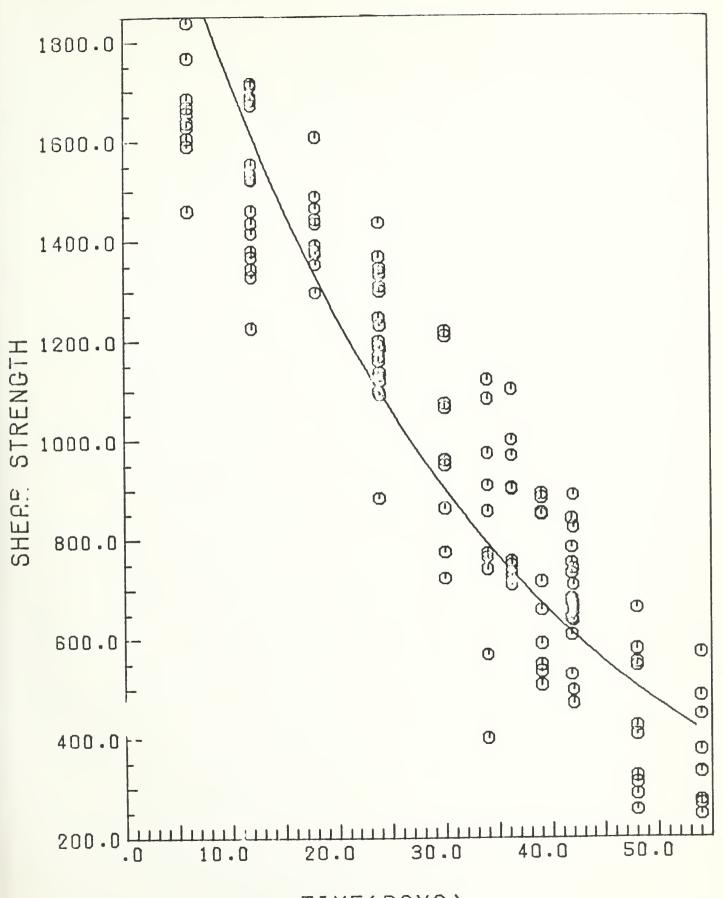
HALF-LIFE AT 20° C.--WET AGING

	Time to	95% Confidence limits		
	half-strength (years)	Lower (years)	Upper (years)	
Wood	400	53	3,000	
Phenolic- bonded	1,100	23	49,000	
Melamine- bonded	6	2	20	





TIME(DAYS)



TIME(DAYS)

## OF SHEAR STRENGTH--WET AGING

Time to half-strength (days)

Aging temperature	Wood	Phenolic- bonded	Melamine- bonded
°C.			
40			279
50			74
60	647	1243	32
67.5			20
70	107	111	
75			7
77.5	74	72	
85	26	29	
100	6	7	

ARRHENIUS PROJECTIONS FOR
HALF-LIFE AT 20° C.--DRY AGING

	Time to	95% Confidence limits		
	Time to half-strength(years)	Lower (years)	Upper (years)	
Wood	1.5 x 10 <sup>6</sup>	8.1 x 10 <sup>3</sup>	2.7 x 10 <sup>8</sup>	
Phenolic- bonded	1.2 × 10 <sup>5</sup>	3.2 x 10 <sup>4</sup>	4.7 x 10 <sup>5</sup>	
Melamine- bonded	1.5 x 10 <sup>5</sup>	8.9 x 10 <sup>3</sup>	1.9 x 10 <sup>6</sup>	

#### BRYAN H. RIVER

Bryan is a Research Forest Products Technologist at the U.S. Forest Products Laboratory, also in the area of adhesive systems and adhesive-bonded products. He received the Bachelor's degree in Forestry in 1962 and the Master's Degree in Wood Utilization in 1969, both from the University of Montana. For the past 9 years at FPL he has specialized in evaluating the durability and mechanical properties of adhesives for wood, and in particular, adhesives for use in building construction.

#### RELATIONSHIP BETWEEN RATE-PROCESS METHOD AND WEATHERING TESTS

Seven different adhesives were evaluated for durability as plywood adhesives by exposing panels and shear test specimens to weathering at the Madison exposure site for nearly 9 years. Wet-strength loss and wood-failure changes were measured as a function of exposure time. The method of exposure accelerated the degradation that would have resulted in most service environments.

Results were compared qualitatively with the information obtained by accelerated aging in the laboratory and analyzing wet-strength loss as a rate process. The rapid initial strength loss that had been observed with acid-catalyzed adhesives on Douglas-fir by dry-heat aging in the laboratory was also detected in their behavior on the test fence. The tendency for some adhesives to hydrolyze during laboratory aging by water soaking was also reflected in test fence performance. A more precise measure of a correlation between test fence results and laboratory-derived data was circumvented by the high variability in the shear strength values.

### RELATIONSHIPS BETWEEN RATE-PROCESS METHOD AND WEATHERING TESTS 1

Ву

ROBERT H. GILLESPIE, Chemist and BRYAN H. RIVER, Technologist

Forest Products Laboratory, Forest Service U.S. Department of Agriculture

How long will a bonded wood product last in service? If the bonded product is a structural building component, normally expected to perform for many years, the answer will likely be an estimate based on an accelerated aging test. This naturally leads to a second question:

How does the response of the bond to the accelerated test compare to its response in a service environment.

The question is simple but the answer is difficult. First the researcher must make concessions to specimen size and geometry for accelerated aging. Second, if he could define "normal service environment," he would have difficulty reproducing it in the laboratory. And last but not least among his problems is the inherent variability of wood.

Presented by Bryan H. River to the 1975 Symposium, "Adhesives for Products from Wood," Wisconsin Center Building, University of Wisconsin, Madison, Wis., September 25, 1975. Will be submitted to Protect Products Journal for consideration to publish.

Maintained at Madison, Wis., in cooperation with the University of Wisconsin.

The purpose of this study was to determine if the differences noted among adhesives in the laboratory-controlled, accelerated aging experiments were related to their performance in outdoor exposure. The general plan for this study was to determine the change in certain joint properties as a function of exposure time in a given environment.

The accelerated aging experiments had been developed to measure rates of shear strength loss and changes in wood failure or locus of failure, viewing these changes as a rate process. Elevated temperatures accelerated the rate of change.

By repeating the exposures at a series of elevated temperatures, it would be possible to establish the temperature dependence of the rate of change in shear strength.

In most cases the log of the rate of change or the degradation

rate will be a straight line relationship with the reciprocal of the absolute exposure temperature. With this relationship established in a relatively short time by using elevated temperatures, it is possible to estimate the rate of degradation at lower service temperatures.

Of the principal degrading forces that commonly act on adhesive joints the accelerated aging environments in this study were designed to evaluate primarily temperature and moisture effects.

Service environments impose thermal, hydrolytic, and swell-shrink effects on exposed material. Naturally the effects vary from one outdoor environment to another.

For the present study, the service environment selected was the Laboratory's outdoor exposure site at Madison, Wis., where the temperature can range from well below freezing to 100° F. Precipitation is frequent with an average total of 30 inches annually.

Plywood was the bonded material chosen for exposure to the accelerated laboratory and outdoor aging environments. Plywood joints in the form of precut shear test specimens were used in the laboratory aging exposures.

Both precut test specimens and 7- by 12-inch panels were placed in the outdoor exposure in 1964, on racks facing south. This selection of unpainted or unprotected specimens and panels accelerated the rate of degradation over what would normally occur if the plywood had a protective coating or partial cover. While this may not represent a typical outdoor exposure it was necessary to do this in order to achieve measurable degradation within 10 years.

The laboratory dry heat aging experiments revealed that all the adhesives tested had resistance to thermal degradation equal to or exceeding either yellow birch or Douglas-fir. Evidence of this was the continually high or increasing amount of wood failure as exposure progressed.

Joints with adhesives requiring an acid catalyst lost strength rapidly during the initial stage of heating, then at a slower rate with prolonged heating. The initial strength loss is the difference between

the original strength and the strength determined by extrapolating the regression equation of the slower rate of strength loss back to zero time. This initial strength loss could be as high as 50 percent as in the case of Douglas-fir bonded with acid catalyzed phenol.

Figure 1 compares the average initial strength loss of joints with neutral or alkaline adhesive systems compared with the average initial strength loss of joints with acidic adhesives systems. The effect was also observed in joints after prolonged storage in a low moisture, normal service temperature environment. Figure 2 shows the same effect in yellow birch joints. In both accelerated aging and storage the effect was most severe in joints of Douglas-fir.

#### Laboratory Water Soak Aging Results

The laboratory water soaking experiments demonstrated the high resistance to hydrolysis of the hot-press phenol, resorcinol, acid catalyzed phenol, and polyvinyl acetate adhesives. These systems were in a class by themselves. The melamine adhesive proved susceptible to hydrolysis, the melamine-urea even more so, and the straight urea hydrolyzed most rapidly.

We can get a feeling for susceptibility to hydrolysis by looking at figure 3, the temperature dependence of the time required for adhesive joints to lose half their original strength. This measure

of durability that we call half-life increases from left to right as the temperature decreases. The highly hydrolysis resistant adhesives are grouped at the top. Wood failure was the typical mode of failure for joints with these systems, so it is reasonable to consider the average curve of these systems to be a good approximation of the temperature dependence of hydrolysis of yellow birch.

#### Outdoor Aging

At various intervals over an 8-year period, five specimens and one panel representing each adhesive-species combination, were removed from the exposure fence for testing. Five specimens were cut from the center of each panel, then the two groups of five specimens were tested wet. In the following graphs each point represents the average value of five specimens tested. The most striking characteristic of the results was the high variability in the wet shear strength data. An example of the variability resulted from testing Douglas-fir specimens cut from the exposed panels.

The results for the hot-pressed phenol adhesive are shown in figure 4 where the percentage shear strength retained, is plotted against the time of exposure. The vertical lines through the data points span the 95 percent confidence limits on the mean. The magnitude and change of these confidence limits are representative of the frustrations attending the analysis of test-fence, rate-process data. One approach

to this problem was to average the total response of the panels to weathering by calculating a least squares regression line using a linear model. Analysis indicated that other models provide a better fit to some of the data. But this linear function, which was also used to analyze the laboratory aging data, still gave an indication of the slope of the response, or degradation rate, and the position of the curves relative to each other. The result of this calculation is this straight sloping line showing the average response to weathering of phenolic-bonded panels over 8 years.

The regression lines for all the adhesives evaluated as exposed Douglas-fir panels are shown in figure 5. The urea resin adhesive lost strength faster than the other adhesives, and there is an indication that melamine-urea was also losing strength faster than the remaining adhesives. However, the loss in strength of the melamine adhesive that might have been predicted from its behavior in the earlier water soaking exposures, was not evident after 8 years of outdoor weathering in panel form. Joints with hot press phenol, resorcinol, and melamine all retained at least 80 percent of their strength after 7 to 8 years of outdoor aging.

There did occur an initial rapid loss of strength early in the exposure of both the acid catalyzed phenol and the catalyzed polyvinyl acetate since the zero time intercept of the regression lines showed only 62 to 68 percent of the original strength. Aside from the urea

and melamine-urea, the Douglas-fir panel bonds seem to respond to the dry
.
heat or thermal effects of the test fence exposure.

The relationships of wood failure to time, for the same series of specimens, shown in figure 6, add emphasis to this indication. The hot pressed phenol, resorcinol, melamine, and catalyzed polyvinyl acetate all had high initial wood failure (85 pct or more) and maintained high wood failure throughout the 8-year exposure period. In other words any strength change in the joint was directly associated with strength change in the wood. The acid catalyzed phenol had low initial but steadily increasing wood failure as exposure progressed. The wood failure-time relationships for these adhesive joints is in substantial agreement with the relationships noted in the laboratory dry heat aging studies.

On the other hand wood failure of joints bonded with urea and melamine-urea declined as exposure progressed. And since we have shown previously that urea and melamine-urea are susceptible to hydrolysis but highly resistant to dry heat thermal effects, it seems probable they are responding to the hydrolytic conditions present in the outdoor exposure.

Results obtained from Douglas-fir plywood that had been exposed as precut shear test specimens were not as variable as those from the exposed panels. Figure 7 shows the linear regression lines for this

series of specimens as shear strength retained versus exposure time.

The most durable joints, those made with hot pressed phenol, resorcinol, catalyzed polyvinyl acetate, and melamine, did not degrade seriously.

Joints with these adhesives maintained 60 to 70 percent of their strength after 8 years of accelerated outdoor aging. Continued high wood failure indicated the loss in joint strength was in the wood rather than the adhesive. The acid catalyzed phenol and the catalyzed polyvinyl acetate bonded joints showed greater initial strength loss just as in the panels aged outdoors and the laboratory dry heat aging. The melamine-urea adhesive lost strength and wood failure declined even more rapidly than in the panel exposures. The urea adhesive in precut shear test specimens failed completely in 4 years of outdoor exposure.

The results obtained with yellow birch were similar to those found with Douglas-fir plywood. Figure 8 shows the linear regression lines for yellow birch plywood exposed as precut shear test specimens. With all the adhesives, degradation was more rapid in yellow birch joints than in Douglas-fir, and the urea and melamine-urea degraded most rapidly on yellow birch just as they did on Douglas-fir. There was one notable exception to the similarity of the behavior between yellow birch plywood and Douglas-fir.

Figures 9 and 10 again show the strength retention in the early stages of exposure to accelerated aging and storage found by extrapolating

the linear regression lines back to zero time and comparing the zero time shear strength to the original shear strength. The strength retained by the joints upon initial exposure to outdoor aging has been added to the figure. In figure 9, the strength retained by Douglas-fir joints with the three acidic adhesive systems, urea, acid phenol, and catalyzed polyvinyl acetate was considerably less than the nonacidic systems in all three exposures.

The acidic and nonacidic systems showed the same relationships in yellow birch joints as in Douglas-fir under both accelerated aging and storage conditions but not under the outdoor aging conditions, as shown in figure 10.

#### Summary

The high variability of the wet strength of plywood exposed to out door aging as panels and precut test specimens, precluded any statistical evaluation of the differences noted. However, trends were observed and related to the results obtained by accelerated aging in the laboratory.

The results confirm that the susceptibility of urea and melamineurea adhesives to hydrolysis govern their performance on the test fence,
and that melamine-urea is much more durable than urea on both Douglas-fir
and yellow birch. The tendency for the straight melamine adhesive to
hydrolyze, that was observed during the accelerated laboratory aging
studies, was not detected during 8 years of outdoor exposure.

Degradation of the wood proved to be the predominate mode of failure in joints with durable adhesives.

The initial rapid strength loss exhibited by acid-catalyzed phenolic and catalyzed polyvinyl acetate adhesives after dry heat accelerated aging, was also observed after weathering Douglas-fir plywood but not yellow birch.

Exposing unprotected plywood panels to weathering at the Madison site accelerated degradation over that normally found in service environments. Significant loss of strength in the wood substrates occurred during the weathering period which would not have taken place were the plywood given at least minimal protection in the service environment.

Generally strength losses were more rapid and extensive when plywood was exposed as precut specimens than when exposed as panels with only the center portion sampled for test. Bond strength losses occurred two to four times faster in specimens than in panels.

The results verified that differences among adhesives that could be determined by accelerated aging in dry heat as well as in water soaking were also observable in their performance during weathering.

## DOUGLAS-FIR

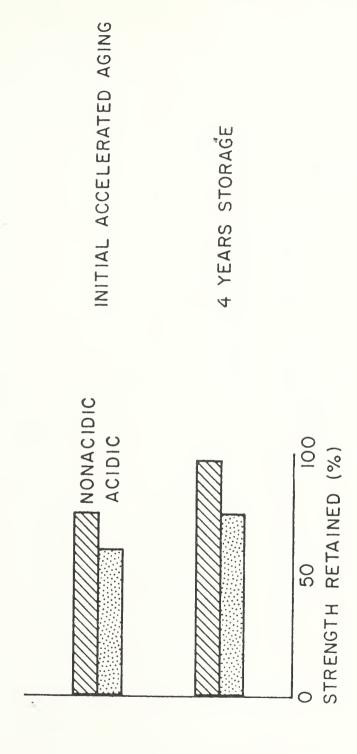


Figure 1.--Initial strength retained by Douglas-fir plywood joints shortly after

exposure was begun to accelerated laboratory dry heat aging and storage at

 $80^{\rm o}$  F, 30 percent relative humidity.

# YELLOW BIRCH

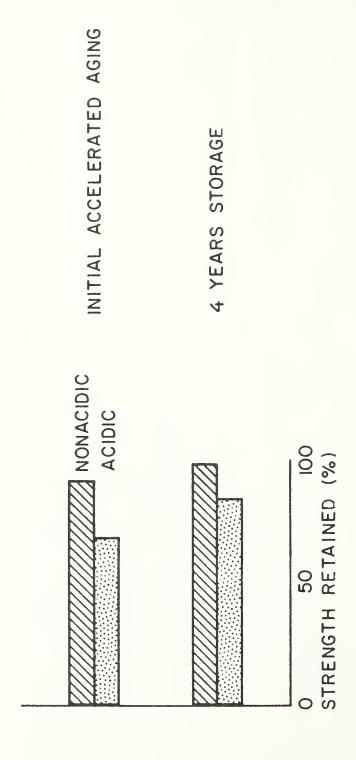


Figure 2.--Initial strength retained by yellow birch plywood joints shortly after exposure was begun to accelerated laboratory dry heat aging and storage at

 $80^{\circ}$  F, 30 percent relative humidity.

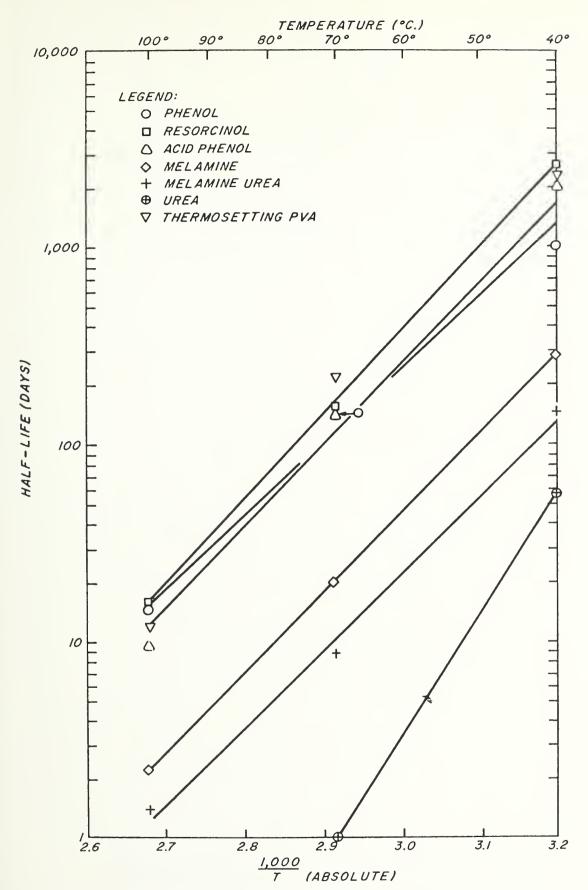


Figure 3.--Temperature dependence of shear strength half-life of yellow birch plywood specimens exposed to water soaking

Figure 4.--Variability of change in wet shear strength with time of exposure

of Douglas-fir plywood panels bonded with a hot-pressed phenol adhesive.

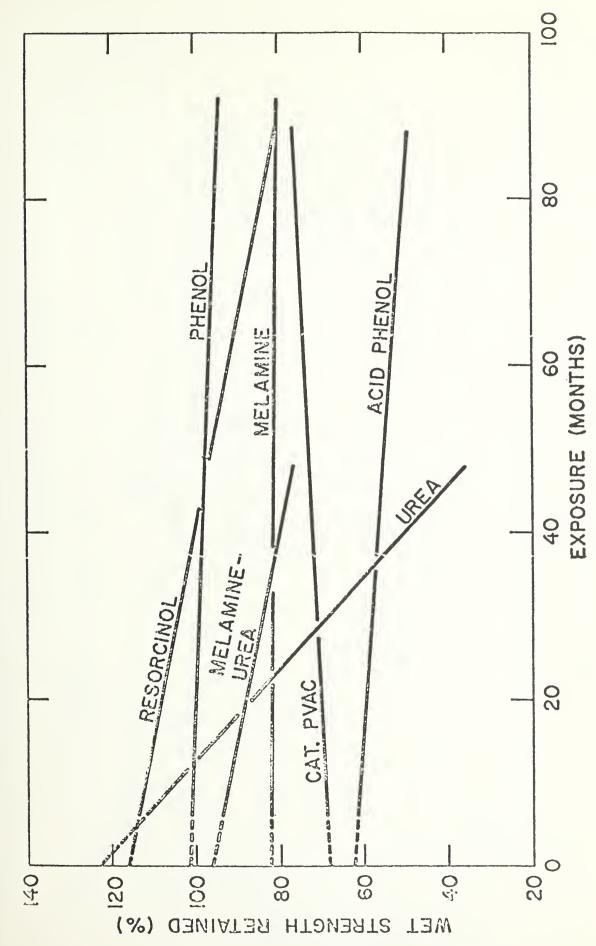


Figure 5.--Linear regression response of the change in wet shear strength with time of exposure of Douglas-fir plywood panels bonded with seven different adhesives.

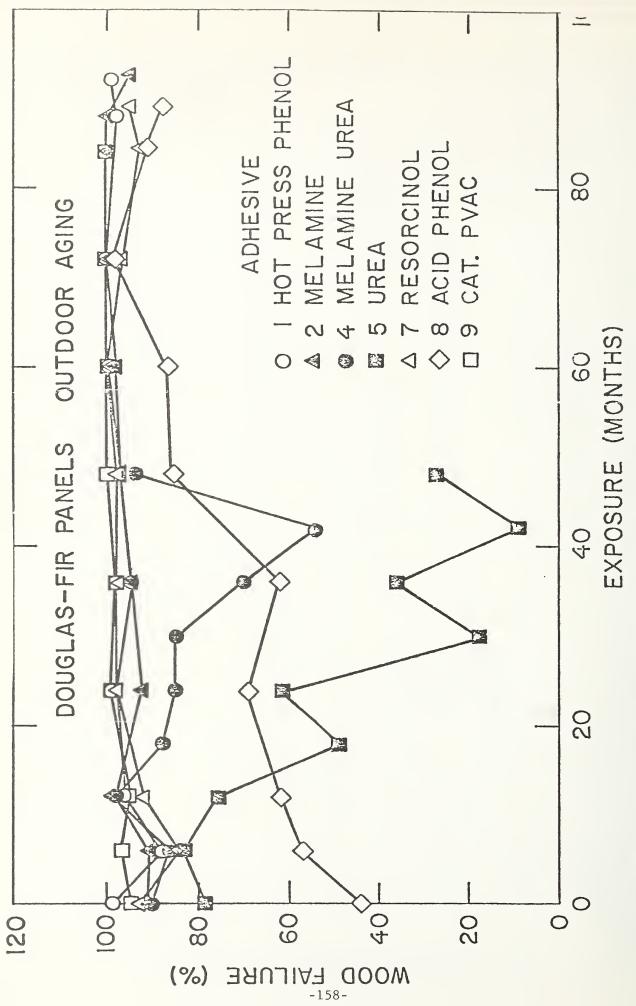


Figure 6. -- Change in wood failure with time of exposure of specimens cut from

Donolas-fir nivrond namels handed with sound different altering

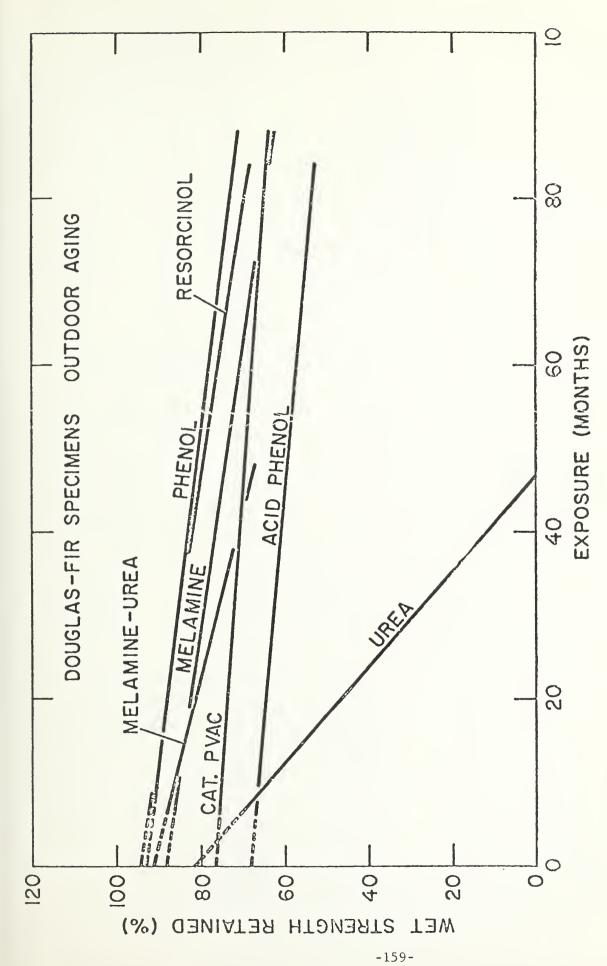


Figure 7.--Change in wet shear strength with time of exposure of precut shear

test specimens from Douglas-fir plywood bonded with seven different

adhesives.

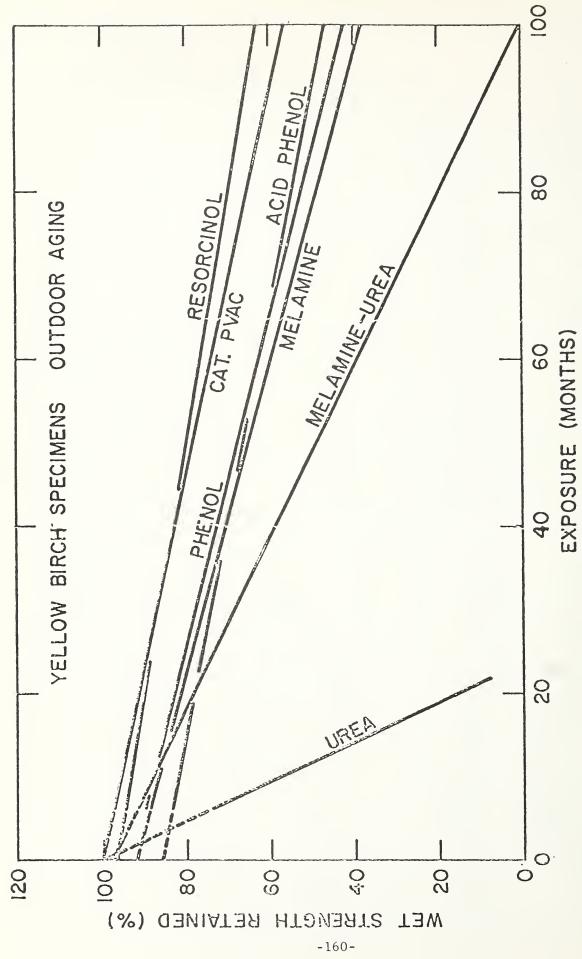


Figure 8.--Change in wet shear strength with time of exposure of precut shear test

specimens from yellow birch plywood bonded with seven different adhesives.

-160-

## DOUGLAS-FIR

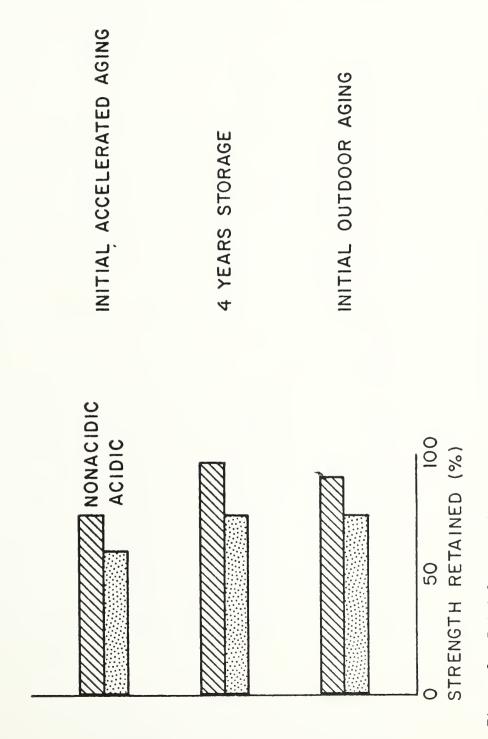
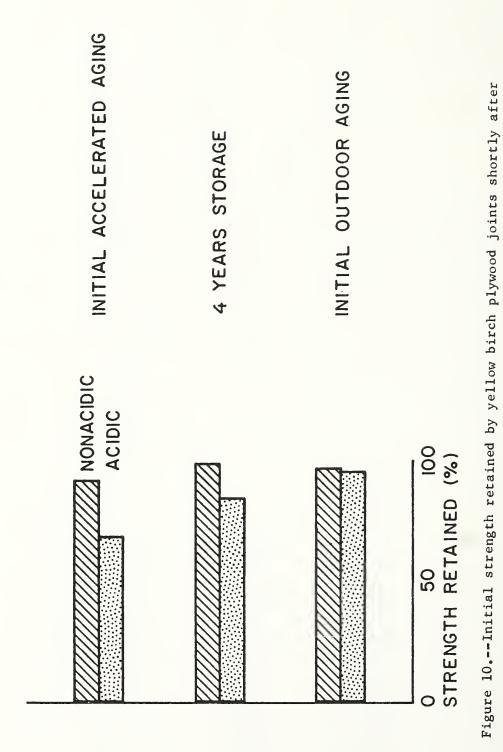


Figure 9.--Initial strength retained by Douglas-fir plywood joints shortly after exposure was begun to accelerated laboratory dry heat aging, storage at  $80^{\circ} \; \mathrm{F}$ ,

30 percent relative humidity, and outdoor weathering.

# YELLOW BIRCH



exposure was begun to accelerated laboratory aging, storage at 80° F, 30 percent relative humidity, and outdoor weathering.

#### ROBERT C. RAYMOND

Bob presently oversees operation of American Plywood Association's eight quality testing laboratories, as well as the gluing research and technical service activities of the Division for Product Approval. He is a Forest Products graduate of the University of Washington, College of Forestry, and has been employed by the American Plywood Association and its predecessor, the Douglas Fir Plywood Association, since 1950. He is a long time member of Steering Committee for the Accelerated Testing of Adhesives and has authored a number of technical and state-of-the-art publications.

#### OUTDOOR WEATHERING OF PLYWOOD AND COMPOSITES

This paper traces the history of durability testing of exterior-type softwood plywood and illustrates correlation with laboratory testing. Included are results of small-scale studies on certain adhesives other than the phenol-formaldehyde types commonly used in exterior softwood plywood.

The development of test procedures for durability testing of plywood/particleboard composite panels is also discussed.

#### OUTDOOR WEATHERING OF PLYWOOD AND COMPOSITES

by
Robert C. Raymond
American Plywood Association

Delivered at the 1975 Symposium "Adhesives for Products from Wood"

Madison, Wisconsin September 24-26, 1975

Until 1938, the glue bond quality of the softwood plywood industry in the U.S. was regulated by a phrase in the Commercial Standard saying "It shall be well manufactured." In the Commercial Standard revision published that year, glueline performance requirements were first established.

Exterior type plywood had been in existence only a little over four years at that time and the industry had little experience with testing it. Fortunately, the U.S. Forest Products Laboratory did have and they had exposed such plywood to weathering when it first became a reality. It was their considered judgement, based on outdoor performance, that, using the now familiar plywood tension-shear test specimen subjected either to the boiling or cold soaking tests, a plywood panel could be considered durable out-of-doors if percent wood failure was at least 50%. Just for how long the panel would remain durable, no one yet knew but the amount of wood failure had been recognized as important by FPL researchers. Don Brouse wrote in 1938: "One of the first impressions gained in this study was the importance of the amount of wood failure developed when testing the joints."

When U.S. Commercial Standard CS 45-38 came out in November 1938, it called for an average of not less than 60% wood failure for each panel with none of the five specimens tested from that panel rating less than 30% wood failure. If 50% wood failure would give good durability after nearly five year's of outdoor exposure, then 60% should be that much better, it was judged.

In the same year, the Douglas Fir Plywood Association started its own outdoor exposure fence near Tacoma, Washington to do its own tests on Exterior plywood durability. Looking backward, we can say that control of location of laboratory test specimens with respect to the piece of plywood actually exposed to weathering was somewhat lacking. The necessity for selecting samples in close proximity to one another and of examining for delamination only those gluelines actually tested in the laboratory became obvious in the early 1940's. Since that time, additions to the exposure fence have been almost entirely of 3-ply specimens 3-1/8"xll" in size cut adjacent to lab shear test specimens. The same piece of face, back and crossband veneer are in both outdoor and lab specimens and veneers are completely free from growth defects.

Today, the exposure fence numbers some 25,000 specimens glued with just about every Exterior plywood adhesive ever approved for use in the softwood plywood industry. All specimens are mounted on a 45° angle facing South to take advantage of maximum exposure to sunlight, rainfall and prevailing winds. Specimens are examined regularly for delamination and a record is kept tying in such delamination with the original wood failure rating of the control tests. Incidentally, shear test loads have always been recorded in control tests too, so that each exposed specimen also has an average load assigned to it.

Perkins, in 1950, first reported the results of correlations of percent wood failure with durability using some 3200 specimens from this exposure fence. The maximum exposure period reported by him was nine years and data included plywood bonded with 28 phenolic adhesives representative of that era. He also reported that he was unable to support any theory that high shear test breaking loads had a bearing on panel durability, although there was evidence to indicate that low loads, below 80 psi, were more often than not associated with low wood failure ratings, and, hence, poor glue bond durability. Conversely, high loads, over 250 psi, were more often than not associated with high wood failure ratings and, hence good durability. I want to stress here that we are talking only about softwood plywood, primarily Douglas fir.

A more recent analysis of wood failure vs. panel glueline durability on some 15,000 matched specimens exposed in the 1950's and 1960's shows the same trends as Perkins' work. These data include a wide variety of softwood species.

Based on these data, minimum wood failure standards for softwood plywood have increased over the years to the point that full Exterior type is required to average no less than 85% wood failure with no more than 10% of the panels rating less than 60% wood failure.

Over the years, many have investigated other approaches to predicting durability of Exterior softwood plywood glue bonds, all of which, even today, are formed with phenol-formaldehyde adhesives. Bryant, in the early 1950's (in unpublished work) found breaking load to be an unreliable predictor except in the very high and very low ranges much as Perkins found. Koch in his exhaustive studies with soughtern pine plywood in the mid-to-late 1960's found percent wood failure to be the best measure of outdoor durability performance of the methods studied.

No one who has studied the question believes percent wood failure to be the <u>perfect</u> method for predicting the durability of softwood plywood bonded with phenolics, but it is certainly the most reliable, fast, laboratory method yet devised.

There is plenty of evidence to indicate that the Exterior boil-shear test using percent wood failure is also a reliable method for predicting durability of phenolic bonds in many imported and domestic hardwoods glued to softwoods or to themselves. Some of the imported woods, however, seemingly do not behave as would be expected, possibly because of their extractive content. A study currently underway at Oregon State University and sponsored in major part by APA aims at determining the best method for repidly predicting bond durability with such species. J.D. Wellons will discuss this question in the afternoon session.

The percent wood failure system of predicting bond durability has been successful with some adhesive systems other than straight phenolics, depending upon the pretreatment of the shear specimens before shearing. A case in point is Intermediate type plywood which may be made with either extended phenolics or heat cured animal blood glues fortified with some phenolic resin. Although virtually no plywood of the IMG type has been produced, exhaustive laboratory and outdoor exposure tests demonstrated the feasability of using % wood failure as a bond durability indicator.

In 1963, A.P.A. made panels in the laboratory with four adhesives to determine whether pecent wood failure could be used to predict outdoor durability. This forerunner of the SCATA work included a catalyzed PVA, a high temperature curing epoxy, an MUF and an Exterior phenolic control. Plywood shear tests were conducted using three pretreatments: the Exterior boil test, cold soaking test and vacuum-pressure test.

The catalyzed PVA showed 100% wood failure in all three tests and fairly high breaking loads as well. There are still, after 12 years, no glueline delaminations over 1/4"x2" in size in any of the exposed specimens but the veneer itself has split in 2/3 of them near the gluelines - probably as a result of the acidic nature of the adhesive. We've observed similar behavior with acid catalyzed phenolics.

The epoxy glued specimens subjected to the boil and cold soaking tests all showed 0% wood failure. In the V-P test, % W.F. ranged from an average of 28% to 80%. In the first seven years of exposure, only one of the nine outdoor specimens had delaminated. After 12 years, five have delaminated extensively and the other four show slight delamination. The 28% specimen is one that shows only slight delamination; the 80% specimens delaminated partially after ten years. Clearly, % W.F. is not a good durability indicator here.

The MUF showed low W.F. ratings in the boil test - 4% to 63% - but good ratings 94% to 100% in the cold soak and V-P tests. The boil test turned out to be a good predictor of durability in this case, with delamination occurring pretty much in the order it indicated. All nine specimens had delaminated at the end of eight years of outdoor exposure.

Control tests on the phenolic showed excellent bond quality in all three procedures and after 12 years there are no delaminations.

So, it appears, the plywood shear test and % wood failure method may <u>not</u> be universally acceptable as a glue bond durability predictor for plywood made with <u>all</u> types of adhesives. This suggests need for a cautious approach to approving Exterior plywood adhesives which contain large quantities of non-phenolic ingredients whether reactive or non-reactive.

A new type of softwood plywood, made with Exterior phenolic adhesive, is about to appear on the scene. Sometimes referred to as a composite panel, it is composed of a veneer face and back with a structural, phenolic bound, reconstituted wood core or crossband. This core may be particleboard, flakeboard or similar board product. The first one to be in full scale production will be Potlatch's Plystran which utilizes oriented wood strand furnish for the core board.

A little over a year ago, APA undertook a study of composite panels, with the cooperation of and partial funding by the U.S. Forest Service, to define strength and other properties of such products. A part of the project was devoted to the development of test methods suitable for predicting outdoor durability and for quality control use. The target durability level was equivalence with softwood G-D sheathing grade plywood bonded with Exterior glue and our ultimate objective was to establish performance requirements using whatever test procedure or procedures evolved as most reliable. The speed with which test results could be obtained was also a consideration. Compounding test method development is the fact that bond durability of such a product has to be concerned not only with the veneer-to-board gluelines but with the bond durability within the board itself. With most boards tested, the latter was, in fact, the limiting factor.

Composite panels for the study were glued in our own laboratory to insure control of veneer quality and gluing conditions. Included in the study were:

- 1. Nine commercially available particleboards four made with urea binder and five with phenolic.
- 2. Two veneer species Douglas fir and southern pine.
- 3. Three types of adhesive Exterior phenolic, extended phenolic and a resin-fortified hot press protein of the IMG type.
- 4. Three-ply and five-ply panel constructions the five-ply used veneer crossbanding.
- 5. Optimum gluing conditions and conditions designed to produce a dried out glueline. These conditions were established through earlier trials.
- 6. Plywood control panels were made with each veneer species and gluing condition to serve as benchmarks in the testing program.

The panels were then cut into test specimens of several sizes and descriptions to provide material for eleven different test procedures plus both permanent and retest specimens to be mounted on the outdoor exposure fence.

Laboratory test procedures included:

- 1. Five plywood shear tests boil, V-P, IMG and ASTM D-1037 accelerated aging pretreatments and dry shear.
- 2. Repeated cycles of the Interior plywood vacuum-soak-dry delamination test.
- 3. A continuous steaming test on 1"x5" specimens.
- 4. And four elevated temperature wet-dry cyclic tests on 1"x5" specimens using delamination over 1/4"x1" in gluelines or boards as the examination criteria.

Results of all test procedures were recorded and correlation with delamination experience on the exposure fence was attempted. Exposure wall examinations have been made every three months.

It was found in prelimary work that plywood shear specimens tested on the 3-ply composite panels had to be kerfed specially in an attempt to force failure to occur at a single glueline. This meant kerfing just through the veneer on the face and almost to the second glueline from the back. Even using this method, however, most shear specimens failed in the particleboard even when veneer-to-board bond quality was known to be of low quality. This was more prevalent in wet specimens than in dry, but occurred more often than not even with dry shears, especially in lower density boards with lower internal bond strength.

The plywood shear test procedures, using % Wood Failure as the bond quality criteria, therefore, appear to be of limited value to predict durability of such composite panels. The boil test, of course, screens out urea binder boards and protein based adhesives. So, of course, did the steaming test, but it was not discriminating of poor phenolic bonds or low durability boards made with pehnolic binders.

Shear specimen breaking load after the various pretreatments was examined as a possible durability predictor. It was a worse indicator than percent wood failure, based on exposure fence results.

The elevated temperature wet-dry cyclic delamination tests appeared to show the best correlation with what was observed to occur outdoors. A number of board failures were occurring, even in three of the phenolic boards after nine months outside, as were delams in composite panels bonded with protein glues and the poorly bonded phenolics.

Refinements were made in the lab cyclic test procedures which improved correlation with outdoor exposure and speeded up the tests. Fortunately, we had provided an adequate number of extra 1"x5" specimens from each panel, matched to the exposed pieces, to accommodate this approach.

The lab test procedure that looks the best to us at this time, after one year of outdoor exposure to compare with, uses two wetting and drying cycles per day. Specimens are placed under 150°F, water and a vacuum of 15" is drawn for 30 minutes, followed by soaking at atmospheric pressure for 30 minutes in the same water. Specimens are then dried at 180°F, in a forced circulation oven for 6 hours. The cycle is then repeated but drying is done overnight for 16 hours. Following this, they are examined for delamination. Delamination at any glueline or within a board over 1/4" deep by 1" in continuous length is recorded, and the procedure is again repeated for as many cycles as desired.

This test combines relative speed - two cycles per day - with what appears to be a desirable level of outdoor correlation.

Outdoor specimens have now been exposed for over one year. Although we are not yet ready to make a firm recommendation for performance requirements for composite Interior plywood panels bonded with Exterior glue, we are tentatively proposing the following; based on five test specimens per panel and twenty panel test lots (100 test specimens):

100% of all test specimens pass 4 cycles (2 days) 90% of all test specimens pass 6 cycles (3 days)

This requirement would assure outdoor durability well in excess of one year.

In addition, we feel the Exterior boil-shear test should be used if only to guarantee that adhesives and board binders will withstand the heat resistance requirements of Product Standard PS 1-74.

In closing, I would like to add only that with the cyclic test requirements proposed, none of the urea binder boards would pass and only two of the five phenolic binder boards used in the study would pass the test requirement.

We expect to continue this study and, in the near future, to expand it to include composites for full Exterior exposure in such applications as siding.

#### SUEZONE CHOW

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#### SOFTENING TEMPERATURE RELATIONS TO ADHESIVE DURABILITY

A method for determining the thermal softening temperature of adhesives will be discussed. Softening temperatures of various wood adhesives will be compared, and application of the technique to the study of the influences of paraformaldehyde addition on phenol-resorcinol resin hardening; the melamine content on urea-formaldehyde resin softening; and the moisture content on softening temperature of casein adhesive. The relationship between the softening temperatures and durability of adhesive panel products will also be discussed.

- Dr. Chow's talk was based largely on information contained in:
- S. Chow, P. R. Steiner, and G. E. Troughton 1975. Thermal Reactions of Phenol-Formaldehyde Resins in Relation to Molar Ratio and Bond Quality. Wood Science 8(1):343-349.
- S. Chow. 1972. Thermal Analysis of Liquid Phenolformaldehyde Resin Curing. Holzforschung 26(6): 229-232.
- S. Chow. 1973. Softening Temperatures and Durability of Wood Adhesives. Holzforschung 27(2):64-68.

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active in the field of wood adhesion.

#### REAPPRAISAL OF DURABILITY OF MELAMINE RESIN ADHESIVES

The influence of cure conditions on the bond durability of a 60/40 melamine-urea resin was studied. With longer cure times and higher temperatures (up to one hour at 140°C) the glass transition point of the polymer was elevated and the bond durability (as measured by accelerated methods) was improved.

### NEW DEVELOPMENTS IN ADHESIVES AND BONDED PRODUCTS

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#### JESSE DAVIS WELLONS III

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#### INFLUENCE OF EXTRACTIVES IN DIPTEROCARP VENEERS ON THE CURE OF PHENOLIC EXTRACTIVES

Difficulties have been encountered in gluing some Southeast Asian woods using phenolic resin. Extractives in veneers from yellow, white, and red meranti (Shorea, Parashorea, and Pentacme spp.), keruing (Dipterocarpus spp.), and kapur (Dryobalanops spp.) were suspected to be able to modify resin cure and to cause inferior gluelines.

The average extractive content for veneers of each trade group was determined by sequential extraction with benzene, ether, ethanol, and water. Yellow meranti had the greatest quantity of extractives (15.6%), followed by kapur (10.1%). In general, ethanol— and water—soluble extractives exceeded other extractives. Extractive migration to the veneer surface seemed to occur with kapur veneers but was insignificant for the other groups.

Ethanol-soluble extractives from all veneers were very acidic and capable of depressing the resin alkalinity significantly. Kapur and yellow meranti were the most acidic woods because of their high extractive content. Adding these alcohol-soluble extractives to a phenolic resin in concentrations similar to those expected to occur in gluelines verified that the acidity of kapur and yellow meranti wood was capable of shortening the resin gel time by 50 percent.

The degree of resin cure was estimated by a spectrophotometric method. In general, ethanol and water solubles of kapur and yellow meranti prevented complete resin cure at much lower concentrations than red meranti and keruing extractives.

We concluded that extractives in white meranti, red meranti, and keruing veneers were unlikely to interfere with the resin-curing process, except by retarding moisture loss from the glueline. However, kapur and yellow meranti extractives were found capable of modifying the curing process in two ways: resin gelation was speeded by the decrease in pH and complete cure was prevented by chemical reaction between the resin and extractives.

#### WERNER CLAD

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#### MODIFIED MELAMINE RESINS FOR PRODUCING PARTICLEBOARD

Phenol resins have been hitherto used in Germany for producing weather-resistant particleboard (Type V 100). This paper deals with tests carried out with melamine resin-bonded particleboards which were to comply with Type V 100. Phenol resin-bonded particle-boards were also tested in order to obtain differentiated ratings and evaluations.

Accelerated exposure tests, outdoor weathering tests, and sustained loading tests, did not give any indication that the melamine resin-bonded particleboards deserved a poorer rating than the phenol resin-bonded boards. In comparison to the melamine resins known hitherto, the modified melamine resin used in the tests has certain advantages, especially with regard to reduced brittleness and increased resistance to moisture.

#### SUMMARY

Phenol resins were hitherto used in Germany for producing weather-resistant particle board (Type V 100). This paper deals with tests carried out with melamine resin-bonded particle boards which were to comply with Type V 100. Comparison tests with phenol resin-bonded particle boards were tested in order to obtain differentiated ratings and evaluations.

Accelerated exposure tests, outdoor weathering tests, and sustained loading tests, which are dealt with in detail, did not give any indication that the melamine resin-bonded particle boards deserved a poorer rating than the phenol resin-bonded boards. In comparison to the melamine resins known hitherto, the modified melamine resin used in the tests has certain advantages, especially with regard to reduced brittleness and increased resistance to moisture.

Key words:

particle board production, melamine resin, accelerated test, outdoor weathering test, sustained loading test.

#### INTRODUCTION

About 40 years ago, urea-formaldehyde glues were used for the first time on a large scale for wood bonding in Germany. The expansion of the application of such glues, of course, entailed demands for improved resin adhesives, special grades, and glues which can be combined with one another.

Since urea-formaldehyde resins can be most readily suited to the requirements imposed, a large range of these resins was developed.

As a result of the increasing amounts of particle board used in the building industry, high resistance to moisture and heat has become an important requirement. Thus, phenol resin glues proved to have unlimited weather resistance. Apart from phenol resins, melamine resins and mixtures of urea resins and melamine and resorcinol resins were also tested. The use of isocyanates as adhesive can be regarded as the latest development.

Investigations into a modification of melamine resin remained unsuccessful for a long time. This project failed on account of the adhesive price, but mainly because of insuperable technical difficulties. Adhesives based on melamine resins were hitherto in no way capable of achieving the durability of phenol resins.

# ACCELERATED AND LONG - TERM TESTS FOR GLUES

distinction is made between efficiency tests and suitability ests. Efficiency tests are carried out with due regard to the practical conditions involved, whereas in the suitability tests, the specimens are often subjected to extreme loads without considering whether these loads are encountered in practice. The more theoretical question as to the absolute bonding performance of a certain adhesive is thus opposed to the more practical question regarding the resistance, durability, and quality of a glue under normal conditions.

Performance and suitability are determined in accelerated and long-term tests. It appears problematic to use the results of an accelerated test alone as a criterion for assessing the behaviour of a bonding in long-term tests.

Fundamental information on this point was given by Gillespie and Lewis (1972). Northcott (1968) reported on 11 different accelerated test methods and stated that none of the investigated methods alone is sufficient for a comprehensive evaluation of the ducability of a glue. Strickler (1968) believes that the development of a universal accelerated test method is more than questionable.

It must be considered that untural elimatic variations result in cyclic leads which often comply in no way with the test conditions employed in the laboratory. The resistance of glues and bondings to againg is normally determined in long-term tests, in

which the change in strength properties is recorded during the entire period. As pointed out by Noack and Frühwald (1973), the resistance of bondings to ageing is of decisive importance for the time during which a building component must retain its full serviceability. According to Kollmann, Clad and Wittmann (1964), long-term trials are necessary because the results thus obtained are essential for applying short-term test values under practical conditions.

The above-mentioned authors also stressed the necessity of using unstandardized tests for examining conditioned or weathered specimens in order to study the effects on the ageing of glue bondings. The increase or decrease of the strength of such bondings as a function of time can be only roughly studied by means of standardized tests. The durability of a bonding cannot be reliably assessed unless the maximum loading limit has been just achieved or exceeded. Thus, additional trials must, therefore, be carried out for this purpose.

# DEVELOPMENT OF BUILDING MATERIALS AND BUILDING REGULATIONS

According to a supplement to DIN 1052 entitled "Holzhäuser in Tafelbauart", wood-based materials can be used for load-bearing and stiffening purposes. The materials must, however, be bonded in such a manner that they can withstand the climatic conditions involved.

Analogously to the plywood standard (DIN 68 705), the plywood standard (DIN 68 763) includes two types of bonding with different resistance to moisture which are bonded with specified adhesives. One type (with lower resistance to moisture) is bonded with urea resins, and the other type (with maximum resistance to moisture) with phenol resins. In an older edition of DIN 68 705, accelerated tests were specified and the type of adhesive to be used was left to the discretion of the manufacturer of wood-based materials.

In view of the present regulations, it is very difficult to achieve new developments in the adhesives field.

DIN 68 763 includes minimum requirements for particle boards for the building industry; it is laid down that such panels (V 100) shall be produced only with adhesives based on phenol or resorcinol resins which cure in alkaline medium. For using a different type of adhesive, one must apply for a special approval by the building authorities and submit the results of extensive tests. This procedure may require several years of preparatory work.

It has also been pointed out in the literature that the system of using specified adhesives for the various standardized wood-based panels (AW 100, V 100) should be examined as soon as possible because the standard does not consider the rapid technical development. In contrast to Germany, where the industry is clinging to this adhesive/wood-based panel coupling system, France and to a certain extent the USA as well have testing standards which do not include any restrictions on the use of existing types of glue. These standards are rather conductive to the development of new systems.

# RESISTANCE OF FORTIFIED UREA AND MELAMINE RESINS

A bonding quality comparable to that of phenol resins was not achieved with any of the large number of commercial types of melamine resin. Carruthers (1952) point of out to a comparatively rapid deterioration of melamine resin-bonded plywood which was exposed in the open in an almost horizontal position. Blomquist (1955) replaced 20 - 100 % of urea resin by melamine resin and 10 - 60 % urea resin by resorcinol resin and thus achieved an increase in durability in cyclic exposure tests and in outdoor weathering tests of plywood. Bondings produced with pure melamine resin lost a considerable proportion of their strength after 48 and 72 hours boiling. Klema (1955) stressed the pronounced ability of melamine resins to harden, and that, for instance, hardened products are insoluble in boiling water. As a disadvantage, he mentioned the danger of overhardening of melamine resins together with increased brittleness. Selbo (1965) reported on trials carried out with specimens which had been stored for more than 19 years in an unheated storage building. The specimens bonded with melamine resin (white oak) showed a decrease in shear strength of 7 %, bondings produced with phenol/resorcinol resins retained their original strength values. In contrast to these examples, bondings of Douglas fir, which had been exposed for 19 years, showed a strength decrease of 13 % for melamine resin, and 17 % for phenol/resorcinol resin.

Although melamine resins have a tendency to become brittle (Witt-mann 1970), they can be readily elastified and modified.

In the trials of Scharfetter (1973), in which specimens were exposed for 5 years under a roof (coastal and inland humidity), bendings produced with urea resins and fortified urea resins (MF) showed approximately the same loss in strength, viz., 14% in comparison to 4% for resorcinol resin.

Finally, melamine/phenol resin moulding compounds should be mentioned in this connection. In comparison to pure phenol resin moulding compounds, these compounds have the advantage that they have lower shrinkage and subsequent shrinkage, and a lower tendency to cracking, especially when exposed to heat and moisture.

One can, however, not conclude from these research results that the durability of melamine resin bondings cannot be improved by modification of the resins. In 1973, Deppe and Erust reported of the first successes in using modified melamine resins, isocyanates and tannin cosins. The requirements imposed on panels bonded with phenol resin could be met with melamine resin only when extremely large proportions of glue are used (12 - 14 %). The same authors indicate a proportion of 10 - 12 % for phonol resin, and in the case of isocyanates, 10 % for the core and 14 % for the outer layer.

For introducing a new adhesive, it is necessary to employ a large proportion of glue in the initial stage in order to be on the same side. This proportion can be reduced after sufficient experience has been gained in the production of panels.

#### TRIALS AND TRIAL RESULTS

A special melamine resin, which was more advantageous than the melamine resins and melamine resin blends (fortified products) known hitherto, especially with regard to reduced brittleness and increased resistance to moisture, was successfully developed by modification. The suitability and performance of the bondings produced with this resin had to be examined and tested. These extensive tests are dealt with below.

In order to achieve a differentiated evaluation, comparison trials were carried out with a modified melamine resin (®Kauramin glue) and a normal commercial grade phenol resin ( Kauresin glue), giving due regard to outdoor exposure tests and sustained loading tests with particle board. Results of the accelerated tests were included in the tables for comparison purposes.

This comparison is of immediate interest because different trends in the production of particle board for the building industry have established themselves in the two European countries with the largest particle board production, viz., in Germany and France. At present, phenol resins are used almost exclusively in Germany according to the standard specifications. Only one manufacturer has received a special permission for using isocyanates. Mainly modified melamine resins are used for the same purposes in France.

#### PRELIMINARY TRIALS

According to Schmidt-Hellerau (1969), particle board bonded with pure melamine resin blended with resorcinol did not yield the expected durability, for instance, in outdoor weathering. Some of the test data are given in Table 1.

Glue mix:

Panel thickness:

100 kg ®Kauramin 650 Powder (melamine resin)

85 kg water

5 kg resorcinol

2.5 kg Hardener 500 Powder

20 kg emulsion

18 mm

8 %

Solid resin on bone-dry chips: Pressing temperature/pressing time: 150 °C/9 min.

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Panel data (average value from 100 individual values)	Immediately after production	After 12 months out- door exposure (45° inclination)
Thickness (mm) Density (kg/m³) Flexural strength (kp/cm²)+ in % of initial value Tensile strength perpendicular to board surface (V 20)	18.0 710 259 100 9.7	18.7 680 199 76 4.3
in % of initial value Tensile strength perpendicular	100	Cracks in outer layer 44
to board surface (V 100) Swelling after 2 hours (%) Swelling after 24 hours (%)	2.1 4.1 11.0	1.5 5.7

Table 1 Testing of particle board produced on industrial scale Results of standard test

Further trials of this type, including tests with pure melamine resins, showed that it would be better to use a larger resin proportion until adequate practical experience has been gained. This ought to yield better strength values in prolonged outdoor exposure tests. Clad and Pommer (1971) have commented in detail on this point. The fact that the species of wood and chip composition play a role as well is regarded as known.

Some of the data of the tests of Schmidt-Hellerau (1969) carried out with particle board bonded with melamine resin are given in Table 2. The data of a similar particle board bonded with phenol resin are given in brackets for comparison purposes.

Immediately after production	After 12 months out- door exposure (45° inclination)
18.1 (18.3) 660 (680)	19.2 (19.3) 630 (640)
323 (353)	299 (334)
10.4 (7.5) 15.5 (20.7)	8.0 (9.3) 20.4 (17.0)
3.1 (3.4) 18.8 (19.5)	3.9 (5.1) 13.9 (15.1)
3.5 (6.4) 9.7 (13.8)	3.5 (3.6)   7.5 (8.7)   21.2 (24.9)
36.1 (67.2)	年.0 (55.7)
	after production  18.1 (18.3) 660 (680)  323 (353)  10.4 (7.5) 15.5 (20.7)  3.1 (3.4) 18.8 (19.5) 3.5 (6.4) 9.7 (13.8) 13.5 (28.8)

Table 2 Testing of single layer laboratory panels Results of standard test

The data indicated refer to a particle board bonded with 12 % of a modified melamine resin (calculated as solids). The data in brackets refer to a conventional particle board bonded with 1 % of phenol resin (calculated as solids).

A special grade of resin designated ©Kauramin 540 Liquid was chosen for further trials for developing a modified melamine resin. Some test values of laboratory panels are given in Table 3 (cf. Schmidt-Hellerau 1969).

#### Glue mix:

Kauramin 540 Liquid (63 %)	100
Emulsion 50 %	. 6.9
Ammonium chloride	1.2
Hexamethylenetetramine	0.5
Water	11.4
Viscosity at 20 °C (mPa·s)	294
Gelling time at 100°C (s)	85
Solid resin on bone-dry chips (%)	11
Solid wax on bone-dry chips (%)	0.6
Moisture content before pressing	11.0

### Panel data (average values from 100 individual values)

Thickness (mm) Density (kg/m²) Flexural strength (kp/cm²) Tensile strength perpendicular to board surface (V 20 kp/cm²) Tensile strength perpendicular to board surface (V 100 kp/cm²) Swelling after 2 hours Swelling after 24 hours Water absorption after 2 hours  Water absorption after 24 hours  Water absorption after 24 hours  27 4		
Tensile strength perpendicular to board surface (V 20 kp/cm²)  Tensile strength perpendicular to board surface (V 100 kp/cm²)  Swelling after 2 hours  Swelling after 24 hours  Water absorption after 2 hours  13.4  5.2  5.2  7.8  11.9	Density (kg/m <sup>3</sup> )	640
Tensile strength perpendicular to board surface (V 100 kp/cm²) 5.2 Swelling after 2 hours 2.8 Swelling after 24 hours 7.8 Water absorption after 2 hours 11.9	Tensile strength perpendicular to	
board surface (V 100 kp/cm²)  Swelling after 2 hours  Swelling after 24 hours  Water absorption after 2 hours  11.9		13.4
Swelling after 2 hours 2.8 Swelling after 24 hours 7.8 Water absorption after 2 hours 11.9		
Swelling after 24 hours 7.8 Water absorption after 2 hours 11.9		5.2
Water absorption after 2 hours 11.9		
	Tief.	· '
	Water absorption after 24 hours	27.4

Table 3 Testing of single layer laboratory panels Results of standard test

The data obtained with particle board produced on an industrial scale are given in Table 4. These data, too, were obtained in comparison trials with melamine and phenol resins.

Panel production:	Type of glue  Modified melamine  resin (Kauramin glue)	Phenol resin (Kauresin glue)
Species of wood Mat forming	Coniferous wood Wind sifted chips Single layer panel	Coniferous wood Poured chips Three-layer panel
Solids resin (%)	12.2	12.0 Outer layer
On bone-dry chips Solid paraffin wax (%) On bone-dry chips Pressing temperature (°C) Pressing time (min.)	1.0 - 180 9.0	(OL) 8.5 Core (C) 1.0 (OL) 1.0 (C) 170 10.5
Panel data: (average value from 50 or 30	individual values)	
Thickness (mm) Density (kg/m <sup>3</sup> ) Flexural strength	36.2 596 222	35·3 553 144
Tensile strength perpendi- cular to board surface (V 20 kp/cm <sup>2</sup> ) Tensile strength perpendi-	5.9	3.4
cular to board surface (V 100 kp/cm²) Evelling after 2 hours (%)	1.9	1.3

Table 4 Testing of single layer and three-layer particle boards Results of standard test

Swelling after 24 hours (%) | 6.1

As can be seen from Table 4, the immediate values of the particle beards produced with Kauramin glue were not poorer than those of the panels bonded with phenol resin.

It must also be considered that high flexural strength cannot be obtained unless the core chips, especially those of low density panels, are not too moist.

No difficulties are encountered in applying the modified melamine resin. An increase of the solid resin proportion of the phenol cosin-bonded particle board would entail a further increase of the chip moisture content so that it would be necessary to prolong the crossing time which anyhow had to be longer than that of the Fouramin glue.

In view of these test results, it appeared very desirable to regard such melacine resin-bonded particle boards as equivalent or even sometice to chench resin-bonded particle boards. But it was not yet possible to assess solely on the basis of the available data whether and under which conditions such panels can be regarded to equivalent to V 100 panels according to the German standards (DTI 68 76), chench resin bonded). The building industry would be ill advised to consider solely on the basis of accolerated test

results that such melamine-bonded particle boards are equivalent to those bonded with phenol resin.

Additional proof must be obtained in outdoor exposure tests and sustained loading tests.

Supplementary tests must be carried out with due consideration of the following points:

The water absorption and the equilibrium moisture content of the panels are lower than those of phenol resin-bonded panels.

The pressing time can be kept as short as that for urea resins.

The fungicidal treatment of such panels requires a smaller amount of active substance than the treatment of panels bonded with highly alkaline phenol resins.

Panels bonded with melamine-urea resins can be rendered flameresistant more readily than panels bonded with phenol resins.

The adhesion of surface coatings on exposure to weathering is much better; the alkali does not effloresce.

Panels of large thickness and low density produced with a mixed condensate have high tensile strength perpendicular to the board surface; difficulties may be encountered in this respect in using phenol resins.

The equipment need not be cleaned between changes from urea-form-aldehyde resins to modified melamine-urea-formaldehyde resins.

Both resins can be freely mixed with one another.

The panels have a light shade; the panels do not turn brown.

In order to assess the resistance of melamine resin-bonded particle board (Kauramin glue) to hydrolysis, flexural strength test specimens were immersed for 2 hours in water at 70 °C, stacked in wet condition in a laboratory drying oven and dried for 24 hours at 105 °C. The individual specimens were tested with a final moisture content of 14 % in the usual way. The phenol resin-bonded particle boards which were included in this test had a final moisture content of 17 %.

The tensile strength perpendicular to the board surface was tested at the same time. The test results are given in Table 5 (ef. Schmidt-Hellerau 1972).

Panel data	Type of glue Modified mel- amine resin (Kauramin glue)	Phenol resin (Kauresin glue)
Solid resin proportion (%)	11	12/8 (OL/C)
before test		
Flexural strength (kp/cm <sup>2</sup> ) Tensile strength perpendicular	295	217
to board surface (kp/cm <sup>2</sup> V 100)	5.6	3.4
after test		
Flexural strength (kp/cm <sup>2</sup> )	209	126
Tensile strength perpendicular to board surface (kp/cm2 V 100)	6.2	3.6

Table 5 Testing of single layer and three-layer panels for industrial purposes

Results of exposure tests

According to these values, the pretreatment of the specimens resulted in a decrease of the flexural strength of approximately 30 % with Kauramin panels and approximately 40 % with Kauresin panels.

Specimens from laboratory panels which had been kept for 6 months in a conditioning chamber at 70 °C and 80 % relative humidity were examined in another test for determining the resistance to hydrolysis.

The test values are given in Table 6.

Panel data (average value from 100 individual values)	Type of glue Modified mel- amine resin (Kauramin glue)	Phenol resin (Kauresin glue)
Proportion of solid resin (%)	12	9
before test		
Tensile strength perpendicular to board surface (V 20 kp/cm <sup>2</sup> ) Tensile strength perpendicular to	11.9	9.6
board surface (V 100 kp/cm <sup>2</sup> ) Tensile strength perpendicular to board surface after 5 hours boiling	5.9	4.7.
(10/0m2)	5.2	4.2
after tost (6 months condition	ing at 70 °c/80 %	· ·)
Tensile strength perpendicular to hoard surface (V 20 kp/cm <sup>2</sup> )	10.6	1.1.4

Table 6 Tasking of single layer panels
Results of exposure tasts

These exposure tests and other tests which are not described in this paper (Schmidt-Hellerau 1974) indicate that the Kauramin panels are at least not inferior to the Kauresin panels even under extreme conditions.

In another connection, Clad (1973) pointed out that the mechanical behaviour of particle boards is dependent on the moisture content.

A moisture content between 10 and 20 % is of particular interest for particle board for the building industry. An evaluation of particle board should always include examinations of the equilibrium moisture content and the moisture distribution in the particle board. The influence of moisture on the change in shape under load is demonstrated by the panels described in Table 4.

Table 7 shows the changes in shape obtained by subjecting specimens of 50 mm x 50 mm x 36 mm to a load of 100 kp for one hour. The values obtained are dependent on the initial moisture content.

Load F kp	Deformation 25/10	n in um at 25/20	25/95
Kauramin panels 100	76 (97 %)	78 (100 %)	259 (332 %)
Kauresin panels 100	58 (72 %)	81 (100 %)	455 (572 %)

Table 7 Testing of single layer and three-layer panels Results of deformation test

As expected, these trials, too, showed that the phenol resinbonded particle board deformed to a great extent under load at high relative humidity.

#### MAIN TRIALS

In addition to the laboratory trials, measures had to be taken to obtain an approval for using the modified melamine resin as adhesive for bonding particle board of Type V 100.

The following three series of tests were carried out for this purpose by an impartial testing institute:

- (a) accelerated tests (laboratory tests)(b) outdoor weathering tests
- (c) sustained loading tests.

These extensive trials are described below.

In this case, too, comparison trials were carried out with a modified melamine resin (Kauramin glue) and a commercial grade phenol resin (Kauresin glue) in order to achieve a differentiated evaluation.

### ACCELERATED TRIALS

The test material used was identical to that described in Table 4. The results obtained in these tests are given in Table 8.

Panel data	MF Kauramin panel	PF Kauresin panel	Number of random samples MF/PF
Thickness (mm) Density (kg/m³) Moisture (%) 20/65	36.20	35.34	30/50
	596	553	30/50
	10.3	9.6	9/15
Swelling in thickness q <sub>2</sub> (%) q <sub>24</sub> Young's modulus in	1.9	5.3 8.0	30/50 30/50
flexure (Mp/cm <sup>2</sup> ) Flexural strength (kp/cm <sup>2</sup> ) Separation strength "top" Separation strength "bottom" Tensile strength perpendi-	34.2	19.7	30/50
	222	144	30/50
	16.8	8.9	30/50
	18.5	8.6	30/50
cular to board surface (kp/cm <sup>2</sup> ) "dry" "V 100"	5.9	3.4	30/50
	1.9	1.3	30/50

Table 8 Accelerated tests
Results of standard tests

Table 9 shows the results obtained in testing the tensile strength perpendicular to the board surface after different pretreatments of the specimens with subsequent reconditioning (R).

		Tensile strength perpendicular to board surface (kp/cm <sup>2</sup> )				
Pretreatment of specimens Exposure and reconditioning	MF Kaura panel	I	PF Kaure panel		Number of random samples MF/PF	
20/65 20/95-20/25+20/65 R 20°/H <sub>2</sub> 0 3 d 20°/H <sub>2</sub> 0 10 d 100°/H <sub>2</sub> 0 2 h 100°/H <sub>2</sub> 0 6 h 100°/H <sub>2</sub> 0 15 h 1 x V 313 + 20/65 R 3 x V 313 + 20/65 R 1 x V 313 + 100°/H <sub>2</sub> 0 3 x V 313 + 100°/H <sub>2</sub> 0 5 x V 313 + 100°/H <sub>2</sub> 0	5.6 5.1 5.9 5.1 2.9 1.5 3.9 2.4 1.3 1.1	(100) (95) (86) (49) (49) (25) (22) (66) (47) (41) (27) (22) (19)	3.4 3.1 - 1.5 1.5 1.3 1.3 1.2 -(2) 0.7 -(2) -(2)	(100) (91) (2) (44) (44) (38) (38) (35) (21)	30/50 15/25 15/- 7-8/20 7-8/20 15/50 15/25 15/- 7-8/25 7-8/- 15/- 15/-	

## (2) not tested

Table 9 Accelerated tests

Test results: tensile strength perpendicular to board surface after different pretreatments followed by reconditioning (R)

The flexural strength, too, was determined after the specimens had been subjected to various pretreatments. The results obtained are given in Table 10.

	Flexu	Flexural strength (kp/cm <sup>2</sup> )				
Fretreatment of specimens Exposure and reconditioning	MF Kaura panel	- 1	PF Kaure panel	sin (%)	Number of random samples MF/PF	
20/65 20/95-20/25 + 20/65 R 20/95 + 20/65 R 1 x V 313 + 20/65 R 3 x V 313 + 20/65 R 5 x V 313 + 20/65 R	222 212 188 171 169 152	(100) (95) (85) (77) (76) (68)	144 133 -(2) -(2) 76 -(2)	(100) (92) (53)	30/50 15/15 12/- 15/- 7-8/15 7-8/-	

Table 10 Accelerated tests

Test results: flexural strength after different pretreatments of the specimens followed by reconditioning (R)

Finally, separation strength tests were carried out with both panels after the specimens had been subjected to different pretreatments. The results obtained are given in Table 11.

Separation strength (kp/cm2) Pretreatment of specimens Exposure and reconditioning PF Number of Kauramin Kauresin random panel samples panel MF/PF (%) (%) (100)8.7 20/65 60/100 17.6 (100)(91) (85) 9.5 -(2) 30/50 30/-20/95-20/25+20/65 R 16.1 (108)20/95+20/65 R 14.9 (85) -(2) $20^{\circ}/H_{0}+20/65 R 3 d$ 15.0 15/-20°/H20+20/65 R 10 d (78)13.8 -(2)15/-(77)100°/f<sub>0</sub>0+20/65 R 2 h 11.8 (67) 6.7 30/100 100°/H<sub>2</sub>0+20/65 R 6 h 100°/H<sub>2</sub>0+20/65 R 15 h 6.3 (72)10.8 (61) 30/50 (62) 30/50 10.9 1 x V 513 + 20/65 R 3 x V 313 + 20/65 R 13.5 (77)-(2)30/-12.7 5.8 15/50 (67)72) 15/- $5 \times V 313 + 20/65 R$ 11.6 -(5)(66)

Table 11 Accelerated tests

Test results: separation strength after different protreatments of the specimens followed by reconditioning (R)

Different pretreating stages are indicated in Tables 9, 10 and 11. The duration of the pretreatment and the exposure conditions were as follows:

- (a) Exposure under moist conditions 20/95 (20 °C/95 % relative humidity): 21 days, followed by reconditioning at 20/65 R
- (b) Cyclic exposure 20/95-20/25: 10 cycles, each 2 x 48 hours (= 40 days), subsequently 20/65 R
- (e) French cyclic test V 313: 1 cycle = 3 days in cold water (20 °C), 1 day frost (-12 °C), 3 days drying (70 °C), tests after 1, 3 and 5 cycles, after reconditioning at 20/65 R
- (d) Temerator in cold water, 20°/H20: 10 days, subsequently 20/65 R
- (e) Boiling Last, 100°/H<sub>2</sub>0: 2 hours, 6 hours, and 15 hours followed by 1 hour cooling in cold water, reconditioning (20/65 R).

The results of the accelerated tests show that both the mechanical and objected properties of the Kauramin panels must be regarded as equivalent to those of the phenol resin-bonded particle boards (Kauresin panels).

#### OUTDOOR EXPOSURE TESTS

Cuttings of the Kauramin panels and Kauresin panels described above were exposed to free weathering for three years. In order to obtain the results more rapidly, the specimens were exposed with an inclination of 45°.

Tables 12 and 13 show the most important test results obtained in the three-year exposure test.

Table 12 shows the results of the tests carried out with the melamine resin-bonded particle boards.

Weathering Properties	(years)	0	0.5	1.0	1.5	2	3
Panel thick Moisture ed Density (kg	ontent (%)	36.20 10.3 596	36.39 10.1 576	36.60 10.7 592	36.63 10.7 566	36.86 11.6 580	36.95 11.2 552
Swelling in thick- ness (%)	$ \begin{array}{ccc} q_{2} & (1) \\ q_{24} & (2) \\ q_{+} & (3) \\ q_{b} & (4) \end{array} $	1.9 6.1 -	1.3 5.2 0.5 0.4	1.3 4.9 1.1 1.0	1.2 4.9 1.2 1.0	1.4 5.2 1.8 1.5	1.7 4.9 2.0 1.2
Flexural st (ko/cm <sup>2</sup> ) (%)	trength	222 (100)	208 (94)	190 (86)	203 (91)	180 (81)	161 (73)
Tensile strength perpendieular to	dry (kp/cm <sup>2</sup> ) (%)	5.9 (100)	6.0 (102)	5.7 (97)	5.5 (93)	5.5 (93)	5.0 (85)
board surface	V 100 (kp/cm <sup>2</sup> ) (%)	1.9 (100)	1.8 (95)	1.8 (95)	1.8 (95)	2.0 (105)	1.7 (89)
Separation strength	"top" (kp/em <sup>2</sup> ) (%)	16.8 (1.00)	15.7 (93)	13.7 (82)	9.7 (58)	11.8 (70)	6.6 (39)
	"bottom" (ko/em <sup>2</sup> ) (%)	18.5 (100)	16.5 (89)	14.0 (76)	12.2 (66)	14.9 (81)	12.0 (65)

(1) (2) Swelling in thickness of the weathered specimens after reconditioning to 20/65.

(5) Irreversible swelling in thickness of specimens for swelling test.

(4) Irreversible swelling in thickness of specimens for flexural test.

Table 12 Outdoor exposure of Knuramin panels (MF)
Results of standard tests

Table 13 shows the values of the phenol resin-bonded particle board (Kauresin panel) for comparison purposes.

Weathering Properties	(years)	0	0.5	1.0	1.5	2	3
Panel thick Moisture con Density (kg	ontent (%)	35.37 9.6 553	35.90 11.5 566	36.02 12.2 561	36.22 12.3 537	36.30 13.1 541	36.46 13.0 534
ness	92 924 94 94 95 96 97 97 98	5.3 8.0	4.4 7.0 1.5 0.8	4.7 6.8 1.8 2.1	4.5 6.3 2.4 3.4	5.4 7.1 2.6 2.2	5.6 6.9 3.1 2.7
Flexural s' (kp/cm²) (%)	trength	144 (1.00)	128 (89)	119 (83)	116 (80)	113 (78)	109 (76)
Tensile strength perpendicular to	dry (kp/cm <sup>2</sup> ) (%)	3. <sup>4</sup> (100)	3.3 (97)	3.2 (94)	2.9 (85)	3.0 (88)	2.8 (82)
board surface	V 100 (kp/cm <sup>2</sup> ) (炎)	1.3 (100)	1.3 (100)	1.2 (92)	1.2 (92)	1.1 (85)	1.1 (85)
Separation strength	"top" (kp/em²) (%)	8.9 (100)	7.2 (81)	7.4 (83)	6.4 (72)	6.6 (74)	4.6 (52)
	"bottom" (kp/cm <sup>2</sup> ) (%)	8.6 (100)	7.6 (88)	8.0 (93)	7.6 (88)	8.8 (102)	7.4 (86)

Results of standard tests (PF)

Even the results obtained in the three-year exposure test do not rive any indication that the Kauramin panel is less durable than the Kauresin panel which was tested at the same time.

The results of the separation tests can be used only with certain restrictions for evaluation purposes because the specifical depth of the milled groove cannot be maintained; this applies especially to specimens which have already been exposed to weathering.

#### LONG-TERM TRIALS

Long-term trials are carried out in order to simulate the practical conditions more closely. In this case, long-term trials were effected to determine the behaviour of particle board under static flexural load. The results of such tests supplement the data on the strength and deformation properties, and give an indication of the durability of the material in practical use.

The tests were carried out with Kauramin panels and Kauresin panels (for details see Table 4).

Three specimens for sustained flexural loading tests were cut out at right angles to the production direction of the panels for each type of adhesive and loading stage.

The width of the specimens was 300 mm, and the distance between the supports in the flexural test was equal to approximately 40 x the panel thickness in test (a) and 33 x the panel thickness in test (b). The length of the test specimens was equal to the distance between the supports plus 80 mm. The central force was applied as a line load over the whole width of the panel. For the long-term trials, the following flexural loads were laid down on the basis of the average flexural strength determined with the panels types (a) and (b) and the flexural strength threshold value of 100 kgf/cm specified in DIN 68 763.

Kaucamin panels 74.0 kgf/cm $^2$  and 33.3 kgf/cm $^2$  Kaucesin panels 48.0 kgf/cm $^2$  and 33.3 kgf/cm $^2$ 

The outdoor weathering tests were subsequently carried out in a test house with two open sides. The swelling and shrinkage of the specimens on moistening was determined with unloaded comparison test specimens (area 630 mm x 300 mm, and 300 mm x 30 mm). The test was carried out over a period of one year. As already mentioned above, the tests were carried out and evaluated by the Forschungs-institut für Holzwerkstoffe und Holzleime, Karlsruhe (cf. Certificate 3637 f/74).

#### TRIAL RESULTS

The most important test results are given in Table 14.

	Type of partic	r Le board	Type o	f le board
Bonding	Kau	enmin.	Кал	resin
(kmf/cm <sup>2</sup> ) Specimen thickness (mm) Density (kg/m <sup>2</sup> ) E <sub>b</sub> (kgf/cm <sup>2</sup> ) f <sub>o</sub> (mm) inttiat deflection	33.3 36.10 591 37000 1.39	74.0 36.14 594 35200 3.22	33.3 35.46 592 20000 1.87	年。6 35、43 590 27390 2、66
ti/to a x 305 days	3.47	3.67	4.17	4.205

Table 14 Data of particle boards

Table 15 contains data on variations in weight and thickness in % ( $\Delta$ G and  $\Delta$ G) calculated on the initial value. Only max.  $\Delta$ G and max.  $\Delta$ G are indicated. These values were determined after approximately 42 weeks - in February 1974 - after a period of comparatively high relative humidity.

	Type of particle board	Type of particle board
Bonding	Kauramin	Kauresin
Changes in weight $\triangle$ G (%), calculated on initial weight		
After 293 days Specimen size 630 mm x 300 mm 300 mm x 30 mm		3.71 5.13
After 365 days Specimen size 030 mm x 300 mm 300 mm x 30 mm		1.91 0.54
Changes in thickness $\Delta$ d (%), calculated on initial thickness		
After 293 days Specimen size 530 mm x 300 mm 300 mm x 30 mm		2.50 2.78
After 365 days Specimen size 630 mm x 300 mm 300 mm x 30 mm	_	1.38 0.50

Table 15 Testing of length and thickness changes of particle boards

The examinations have shown that both the absolute deflection yalues and the croep factors of the Kauramin panels are lower than those of the Kauresin samels.

The maximum noisture absorption of the Kaucamin ennels was 2.85 % and what of Kaucamin conels 3.71 %. The thickness swelling values were 1.72 % and 2.50 % respectively.

The difference in the behaviour of the two panels on exposure to moisture increases with decreasing specimens size.

The 700 mm is 50 mm nore imens showed a regrission of 3.77 % and 3.15 %. The specimen thickness changed by 2.21 % and 2.76 %.

despositing these facts, it can be said that the sustained flexural today test values of the Kauresin panels exposed to catural well-design conditions were in no case infector to those of the Kauresin panels.

The better long-term test values of Kauramin panels in comparison to those of Kauresin panels can be regarded as adequately reliable. It must be mentioned in this connection that the creep factors of the Kauramin panels determined in the various loading stages were lower than those of the Kauresin panels.

#### CONCLUSION

It is obvious that the most hygroscopic panels showed the largest deformation in the long-term loading tests. As the trials have shown, these values were obtained with the phenol resin-bonded panels whose strength properties, in spite of this deficiency, have never caused any complaints. Kauramin panels showed higher flexural strength. This must be regarded as a positive factor especially when the panels are used in the building industry.

The results of the accelerated tests and the outdoor exposure tests, which have been carried out for more than 3 years, too, do not give any indication that panels bonded with Kauramin should be given a lower rating than panels bonded with phenol resins.

Melamine resin-bonded particle boards have already proved themselves in the building industry in France, where no restrictions are imposed on the type of glue to be used for particle board.

After the approval of particle board bonded with melamine resins (cf. DOS) in Germany, practical experience will show whether the two panel types should be given the same rating or whether preference should be given to panels whose absorptivity is similar to that of the wood.

#### SUMMARY

After the development of a special melamine resin for wood bonding, which was superior to all the other melamine resins and melamine resin blends especially with regard to reduced brittleness and increased resistance to moisture, it was necessary to test the resultant bondings for their suitability and performance. These tests are dealt with in this paper. In order to achieve a differentiated rating and evaluation, comparison trials were carried out with a melamine resin (Kauramin glue) and a commercial grade phenol resin (Kauresin glue). The results of the accelerated tests, outdoor weathering tests and sustained loading tests carried out with particle boards did not give any indication that the panels bonded with Kauramin should be given a lower rating than phenol resin-bonded panels.

The tests carried out, of course, are of great significance for a special approval by the Building Authorities, for instance, for the inclusion of Kauramin-bonded particle board into Wood-Based Material Class 100. The following is of importance for this purpose:

According to the Supplement "Holzhäuger in Tafelbauart" to DIN 1052, wood-based materials for load-bearing and stiffening ourposes must be bonded in such a manner that they are capable of withstanding the climatic conditions involved. Analogously to the quality standard for plywood for the building industry (DIN 68 705, Blatt 3), the quality standard for particle board for the building industry (DIN 67 763) includes two bondings with different resistance to moisture. Phenol resin, whose durability is known from the production of plywood, is specified as adhesive for wood-based materials of Class 100. Other types of adhesive which are to be used for wood-based materials of Class 100 cannot be readily tested beforehand for their resistance to weathering by the accelerated tests which were developed for phenol resins. A special approval by the Building Authorities must, therefore, be applied for these resins and the wood-based materials bonded with them. Building materials which do not comply to standard with regard to the adhesive used cannot be officially approved unless their usability can be proved by test certificates, which in turn are based on extensive suitability and performance tests. The tests carried out with a new modified melamine resin are described in this paper.

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After receiving his Ph. D. in Chemistry from the University of Delaware in 1959, Dr. Garrett worked for several companies developing both structural and non-structural adhesives. His past activities have included development of polyesters, phenolics, epoxy adhesives, molding compounds, tire cord adhesives, glass fabric and graphite filament prepreg, core-to-skin adhesives, and metal bonding adhesive. As Chief Chemist and Manager of Research and Development for Roberts Consolidated Industries the past several years, he has been heavily involved in the development of new high-performance contact, construction, and carpet adhesives.

#### DEVELOPMENTS IN CONTACT ADHESIVES

Contact adhesives are widely used in factory and home because of their ease of application, adhesion to a variety of surfaces, and rapid development of handling strength. These versatile adhesives are designed for application by spray, brush, roll or curtain-coating equipment from either organic solvent or water-based systems.

While the largest quantities of contact adhesives are currently sold in organic solvents, the demand for water-based types has been increasing, primarily because of safety and ecological consideration. Recent improvements in neoprene latex contact adhesives have brought cost/performance factors more in line with solvent types, providing additional impetus for using water-based contact adhesives.

In this paper, the latest developments in both solvent and water-based contact adhesives are described in detail. Mechanical properties and application methods are compared, and the effects of adhesive loading, open time, adhesive pattern, bonding temperature, and other parameters on performance of the adhesives are discussed.

#### DEVELOPMENTS IN CONTACT ADHESIVES

By B. R. Garrett

#### INTRODUCTION:

The use of contact adhesives has grown remarkably over the past decade. Characteristics of these adhesives that undoubtedly have contributed to this growth are their ease of application, versatility in bonding to a variety of materials and immediate development of strength once a bond has been formed. In this paper, the characteristics of the various contact adhesives used in the manufacture of laminated doors, sandwich panels, counter tops, kitchen cabinets, interior wall partitions and other fabricated plastic products will be discussed.

Most contact adhesives for these applications are based on neoprene rubber and are formulated for application by spray, brush, rollcoating or curtain-coating. The standard bonding procedure is to apply the contact adhesive to both bonding surfaces, air dry or oven dry and combine under pressure rolls using enough pressure (usually 40-70 psi) to assure good contact.

Materials bonded in this manner develop handling strength immediately and are ready to be post-formed, routed, stamped, drilled or sawed as needed. This characteristic of immediate strength development makes contact adhesives extremely well suited for home use as well as for high speed industrial laminating and forming operations.

Bonding versatility is another outstanding quality of contact adhesives. Porous and non-porous materials, flexible and rigid substrates, rough or smooth surfaces can all be bonded by selection of the proper contact adhesive. Typical applications include:

- 1. High and low pressure plastic laminates bonded to aluminum, steel, plywood or particleboard.
- 2. Polyurethane and polystyrene foams bonded to themselves or to wood, metal or plastic.
- 3. Rubber, leather, fabric, paper, cork and felt can be bonded to themselves or to other flexible materials.

It is this bonding capability, combined with easy application, dependability and general usefulness that has made neoprene contact adhesives so popular. In recent years, several non-neoprene contact adhesives have been marketed for industrial uses. Most of these are block SBR types and, while of lower quality than the neoprenes, one or two appear to be satisfactory for manufacture of flat laminated products.

#### ADHESIVE COMPOUNDING CONSIDERATIONS:

Neoprene elastomers quickly found their way into contact adhesive because of their high tensile strength and good crystallization characteristics, producing quick grab and rapid development of cohesive strength in the adhesive without curing. There are many neoprene elastomers of different chemical structures, in several molecular weight ranges, available to the adhesive compounder. Using these elastomers, phenolic resins, metal oxides, tackifiers with suitable solvents, he is able to control rheological properties of adhesives, extend open time and tack range and adjust viscosity.

A good contact adhesive should have the following properties:

#### As Formulated

Easy application
Rapid drying
2-3 HR open time
Quick grab
Rapid strength build-up
Minimum settling
Freeze/thaw resistance
Non-phasing
6-12 month shelf stability

#### As Film

Adhesion to many surfaces
Toughness and flexibility
Good shear and peel strengths
Resistance to aging and weathering,
moisture, heat

Not only must the chemist formulate the adhesive to the above properties, he must also give careful consideration to:

- 1. EPA, OSHA, DOT regulations and local ordinances;
- 2. the application method and specific equipment used by the fabricator;
- 3. the selling price of the finished adhesive.

In attempting to meet all these requirements, it is no wonder that most contact adhesives are complex, carefully balanced formulations.

#### DEVELOPMENTS IN CONTACT ADHESIVES:

There are many different type neoprene contact adhesives available to fill the varied needs of the consumer, industrial and trade sales markets. Some of the more frequently used types are shown in FIGURE 1 with typical peel and shear properties in FIGURE 2. The most popular adhesives have been the basic general purpose spray and brush grades because of greater utility and lower cost. Various modifications of this type are being sold, some with slightly higher peel strength or quicker grab but, still a general purpose grade.

Usage of non-flammable adhesives has not been as great as the other types primarily due to the inherently higher selling price. Sales of the non-flammable grades will undoubtedly increase if OSHA and EPA restrictions on the use of solvents becomes more rigid.

High speed post-forming lines require a quick drying, high strength, heat-resistant sprayable contact adhesive, preferably one that sprays a wide, uniform fan. There are not many of these produced. ROBERTS 2051 is particularly well-formulated combining all of the above properties with a uniform droplet spray pattern and better coverage per gallon than with general purpose types.

The selling price for higher performance contacts is naturally higher than prices for general purpose grades. Analysis of coverage figures has shown that cost per square foot for adhesive bonded materials frequently favors the high performance adhesives. About 2.5-3.5 grams of adhesive solids are coated on each bonding surface from solvent-based contacts, depending upon the application. Even at today's adhesive prices, this coverage costs the fabricator only 1-1/2¢ to 1-3/4¢ per square foot of bonded product.

During the past couple of years, interest in water-based neoprene contact adhesives has gradually increased as compounding technology improved and more efficient application equipment became available. The need to conform to updated OSHA, EPA and DOT regulations has provided additional impetus for conversion to water-based adhesives.

Water-based contact adhesives offer many advantages to the laminator:

#### Advantages

Wide range of solids
Wide viscosity range
Non-flammability
Easy application by spray,
brush or roll
Low volatility
Easy clean-up
Can dry at reasonable rate
Continuous application is
possible
No waste solvent

#### Disadvantages

Lower water resistance
Lower resistance to freezing
and thawing
Slow drying on non-porous surface
Requires lined or plastic containers

Overall quality of neoprene latex contact adhesives has improved; peel strengths are higher and high temperature resistance has increased. Adhesive costs to the fabricator are essentially the same as for solvent-based contacts, 1-1/2 ¢ to 1-3/4 ¢ per square foot of bonded product, although about 10% more adhesive is needed to get equivalent performance.

#### APPLICATION EQUIPMENT:

Equipment manufacturers have done an outstanding job in designing equipment to properly handle water-based adhesives. New spraying techniques give better break-up of latex adhesive droplets, producing more uniform deposition. More efficient drying ovens are now being used in coating lines and water-based contact adhesives can be dried more rapidly with about the same overall energy requirement as for solvent-based contacts.

One equipment manufacturer, Bechtold Engineering, has developed automatic spray lines complete with conveyor, spray booth, transverse spray unit and drying tunnel for solvent (Model AFG-60) and water-based (Model AFG-W) adhesive applications. In addition to their Mark 9 Roll-A-Matic post-forming machinery, they recently added a lower cost Mini-Line which includes hot spray, indexing table, pinch roller, mini-Ram and coving machine. With this new equipment, it is now possible to manufacture fabricated plastic products in long, continuous production runs using water-based contact adhesives.

Additional innovations in equipment design can be expected from equipment suppliers to assist fabricators in making the transition from solvent-based to water-based contact adhesives.

A typical manufacturing layout used by many manufacturers of wall panels, doors, counter tops and cabinets is shown in FIGURE 3. The modular design concept permits easy adjustment to other plant configurations.

#### FUTURE EXPECTATIONS:

Over the next few years, adhesive bonding of fabricated plastic products with contact adhesives is expected to increase. The current trend towards faster drying adhesives with improved green strength will undoubtedly continue. SBR, acrylic and other elastomers should find increasingly greater usage, particularly in water-based contacts.

Rising costs, the need for greater conservation of energy and raw material sources are expected to accelerate the transition to water-based contact adhesives. Naturally, if OSHA or EPA find it necessary to increase restrictions on the use of solvents, the conversion to water-based contacts will be even more rapid.

Manufacturers of fabricated plastic products will continue to search for ways to combat rising material and labor costs. This need will no doubt generate improved application equipment and more effective adhesive bonding systems. Emphasis will continue to be on increased production rates and lower unit manufacturing costs. To achieve these goals will require close cooperation between fabricator, equipment design engineer and the adhesive formulator.

# FIGURE 1

TYPE	CONTACT ADHESIVE	CHARACTERISTICS	BONDING APPLICATIONS
I	General Purpose Spray	Moderate fan width (12") ) 1-3 hr. open time ) Good shear and peel ) 18-20% solids )	Cold or hot spray laminating and post-forming.
II	Non-Flammable Spray	Similar to Type I but ) in non-flammable solvent )	Bonds metals, woods, laminates, foams, honeycomb.
III	Heat Resistant Spray	Wide fan width (18")  Fast drying  1/2-1 hr. open time  20-23% solids  Good shear, high peel  High heat resistance	Cold or hot spray High speed laminating and post-forming. Bonds metals, woods, honeycomb, foams, drywall, laminate.
IV	High Performance Rollcoat	1-2 hr. open time 19-22% solids Good shear and peel Good heat resistance Slower strength buildup	Rollcoat and cold spray Honeycomb laminating of doors and panels.
V	General Purpose Brush	Easy brushability ) 3 hr. open time ) Good shear and peel Rapid drying rate.	Brush or roller laminating and post- forming. Bonds metal, woods, laminates, core
VI	Non-Flammable Brush	Similar to Type V but in non-flammable solvent )	materials.  Home applications.
VII	Water-Base General Purpose	20-40 min. dry time 1/2-1 hr. open time  '45-55% solids Available in several viscosities. Good shear and peel Non-flammable  '' )	Spray, brush, rollcoat, curtain coat laminating and post-forming. General bonding home applications. Bonds laminates, woods plaster, hardwood, veneers.

FIGURE 2

### TYPICAL STRENGTH VALUES

CONTACT ADHESIVES:	<u>I</u>	<u>II</u>	III	IV	<u>V</u>	<u>IV</u>	<u> VII</u>
PROPERTIES -							
Peel Strength, piw (Canvas to H.P.L.)*							
16 hours	12	10	15	10	15	12	12
7 days	20	15	23	15	26	15	17
14 days @ 120°F - Test R.T.	15	15	17	20	21	25	25
Shear Strength, psi (Plywood/H.P.L.)*							
1/2 hour	200	150	150	175	125	100	75
l day	250	200	250	225	150	125	250
7 days	325	250	300	275	250	300	350
7 days @ R.T Test @ 240°F	15	10	22	17	12	12	20

<sup>\*</sup>H.P.L. = High Pressure Decorative Laminate

က

FIGURE

7-29-69

Roll-A-Matic

Conveyor

-204-

#### WALTER C. KANIA

Walter is Special Projects Manager for National Adhesives, Division of National Starch and Chemical Company of Bridgewater, N.J. He received his B.A. in chemistry from New York University and his M.S. from the Newark College of Engineering. He has spent over 20 years in the chemical field, the major portion in the development, technical service, and marketing of adhesives used in the wood product and related industries.

## DEVELOPMENTS IN HOT MELTS, EMULSION, AND SOLVENT SYSTEMS

Hot melts, emulsion, and solvent adhesive systems account for a major portion of the adhesives used in the wood products industry.

Significant strides have been made toward more effective utilization of the unique advantages each of these systems can offer. Advantages, disadvantages, and changing needs will be reviewed and how these have led to the development of new products and end uses.

The rapid bond formation characteristic of hot melt adhesives has accelerated their use, leading to technological advances to meet more demanding end-use requirements. Major improvements in heat-resistant capabilities have resulted in commercial utilization in areas such as large surface area bonding and improved sandability. Specific physical properties and end-use examples will be discussed.

Advent of crosslinking emulsions some years ago to include exterioruse conditions has been expanded to include wood metal combinations. Operational methods, adhesion, performance data, and examples of use will be described.

The trends occurring in solvent adhesives systems will be examined as they relate to current environmental and safety conditions, and how these are affecting current developmental activity and new products.

#### EDWARD R. HARRELL

Ed is Technical Supervisor, Sealants and Coatings Laboratory, for the 3-M Company. He graduated from Illinois College in 1957 with a B.S. degree in Chemistry. He then spent 2 years in graduate study of organic chemistry at Indiana University before joining the 3M.Company as a product development chemist in the Structural Adhesives Group of the Adhesives, Coatings and Sealers Division.

Since 1962, Ed has been a member of the Sealants and Coatings Laboratory group of this same division. He has been instrumental in the development of a number of polyurethane-based sealers, coatings, and adhesives used in both the construction and industrial markets.

#### DEVELOPMENTS IN POLYURETHANE-TYPE ADHESIVES

Polyurethane-type adhesives have certain properties that make them particularly suited to the bonding of wooden assemblies. New products are being developed which display unique characteristics in terms of cure rates and ultimate performance.

One-part, fast-curing urethane systems, some of which exhibit "wood tearing" strengths in less than 15 minutes at 75°F will be discussed. High strength, tough, flexible, durable bonds are possible using this technology.

Two-part, structural strength, urethane-type adhesives will also be discussed. New prototype formulations have been developed which display excellent heat and water-soak resistance which heretofore were areas of weakness in urethane adhesives.

Both the one- and two-part systems cure rapidly at room temperature and require little or no pressure during the bonding process, thus potentially reducing bonding costs. Some of these systems have excellent gap-filling characteristics.

## DEVELOPMENTS IN POLYURETHANE-TYPE ADHESIVES

BY

### EDWARD R. HARRELL - 3M COMPANY

Polyurethane type adhesives have certain properties that make them particularly suited to the bonding of wooden assemblies.

New products are being developed which display unique characteristics in terms of cure rates and ultimate performance.

One-part, fast-curing urethane systems, some of which exhibit "wood tearing" strengths in less than fifteen minutes at 75°F will be discussed. High strength, tough, flexible, durable bonds are possible using this technology.

Two-part, structural strength, urethane-type adhesives will also be discussed. New prototype formulations have been developed which display excellent heat and water soak resistance which deretofore were areas of weakness in urethane adhesives.

BOTH THE ONE AND TWO-PART SYSTEMS MENTIONED ABOVE CURE RAPIDLY AT ROOM. TEMPERATURE AND REQUIRE LITTLE OR NO PRESSURE DURING THE BONDING PROCESS. THUS POTENTIALLY REDUCING BONDING COSTS. SOME OF THESE SYSTEMS HAVE EXCELLENT GAP-FILLING CHARACTERISTICS.

# INTRODUCTION

ADHESIVES AND SEALERS TO THE INDUSTRIAL AND CONSTRUCTION MARKET FOR SEVERAL YEARS.

THESE PRODUCTS HAVE ENJOYED WIDE ACCEPTANCE NOT ONLY DUE TO THEIR GOOD PERFORMANCE PROPERTIES, BUT BECAUSE OF THEIR SUITABILITY FOR ON-SITE APPLICATION UNDER A WIDE VARIETY OF CONDITIONS.

THESE POLYURETHANE PRODUCTS ARE PARTICULARLY SUITED TO THE BONDING OF WOODEN ASSEMBLIES SINCE THEY ACTUALLY REACT CHEMICALLY WITH WOOD SURFACES TO FORM VERY STRONG BONDS.

One of our standard products is approved against the American Plywood Association AFGO1 specification covering glue-nail flooring uses. There also is an International Congress of Building Officials research recommendation written around the use of this product as the adhesive in a Laminated Roof-Decking configuration.

THE EVER INCREASING DEMAND FOR BONDING PROCESS AND PRODUCTS THAT SAVE
TIME AND ENERGY HAS LED US TO EXPLORE THE AREA OF FAST, ROOM
TEMPERATURE CURING ONE AND TWO-PART WOOD ADHESIVE SYSTEMS.
THE FIRST TWO NEW PROTOTYPE PRODUCTS DESCUSSED ARE ONE-PART
SYSTEMS.

# NEW PRODUCTS - ONE PART

TECHNOLOGY - URETHANE, MOISTURE CURING.

CURE CONDITIONS - AMBIENT - NO HEAT REQUIRED AND ONLY CONTACT PRESSURE REQUIRED.

# TABLE 1

PHYSICAL PROPERTIES OF LIQUID	PRODUCT A	PRODUCT B
COLOR	CLEAR AMBER	LIGHT TAN
% SOLIDS	50	88
PASS RULE 66	YES	YES
OPEN TIME (MAX.)	3 MIN.	25 MIN.
FORM	JELLY-LIKE	GUN GRADE
	PASTE.	THIXOTROPIC
		PASTE.
FLASH POINT	50°F	OVER 150°F
		(TOC)
PHYSICAL PROPERTIES - CURED FILM		
TENSILE	4600 PSI	2000 psi
ELONGATION	10%	150%

THE FOLLOWING TABLE ILLUSTRATES THE CURE RATE AND STRENGTH PROPERTIES

OF ONE OF OUR STANDARD PRODUCTS VERSUS THESE TWO NEW PROTOTYPE

ONE-PART PRODUCTS.

TABLE 11

	CURE RATE AND ULTIMATE STRENGTH	STRENGTH	
	SCOTCH GRIP BRAND MOOD ADHESIVE 5230	PRODUCT A	PRODUCT E
TIME TO REACH 100 PSI:	20 - 24 HRS.	4 MINUTES	30′ - 1 HP.
TIME TO REACH 400 PS::		8-12 MIN.	1-1/2 - 2 HPC
ULTIMATE STRENGTH: TIME TO ACHIEVE:	7 DAYS	24 HOURS	24 HOURS
VALUE (PSI)			
AT 75°F	250-350	*700-1100	*700-1000
AT 160°F	and the state of t	**626	* 568

ABOVE RATE OF STRENGTH BUILDUP DATA WAS OBTAINED ON DOUGLAS FIR TO DOUGLAS FIR 1" WIDE OVERLAP SHEAR SPECIMENS USING A 2-4 MIL GLUELINE CURED AT 77°F/50% RELATIVE HUMIDITY. THE BONDS WERE PULLED AT A 2"/MINUTE JAW SEPARATION RATE ON AN INSTRON TESTER. NOTE

<sup>\*</sup>RESULTS VARY WIDELY DEPENDING UPON WOOD GRAIN STRUCTURE.

<sup>\*\*</sup>PASED ON ONLY ONE SET OF SPECIMENS

THESE NEW ONE PARTS EXHIBIT GOOD ENVIRONMENTAL RESISTANCE AS INDICATED BY THE RESULTS LISTED IN TABLES III AND IV.

# TABLE III TEST RESULTS VERSUS AITC-110 CYCLIC DELAMINATION TEST

WOOD - DOUGLAS FIR TO DOUGLAS FIR.

TEST SPECIMEN - 6 INCHES LONG, 3 INCHES WIDE X 3/4 INCHES THICK.

#### PROCEDURE:

- A. IMMERSE SPECIMENS IN 65-85°F WATER IN PRESSURE VESSEL.
- B. DRAW A 20-25" OF MERCURY VACUUM AND HOLD FOR 30 MINUTES.
- C. RELEASE VACUUM AND APPLY A POSITIVE PRESSURE OF 75 ± 5 ps: FOR TWO HOURS.
- D. REMOVE SPECIMENS AND DRY FOR 10 HOURS AT 160°F AND A RELATIVE HUMIDITY OF 8-10%.
- E. OBSERVE DELAMINATION MUST NOT EXCEED 5%.
- F. REPEAT CYCLE

#### RESULTS:

PRODUCT A (UNFILLED) PRODUCT B (FILLED)

PASSES 2 CYCLES - PASSES 4 CYCLES - FALLS AFTER 3RD CYCLE FALLS 5TH CYCLE

# RESISTANCE OF ADHLSIVES TO CYCLIC LABORATORY AGING CONDITIONS (ASTM D-1183-70 PROCEDURE C "MODIFIED")

## CYCLL:

PERIOD IN HOURS	TEMPERATURE, °F	RELATIVE HUMIDITY, %
48	160 ± 5	10
48	73.4 ± 2	IMMERSED IN WATER
8	-40	ABOUT 100
64	$100 \pm 3.5$	ABOUT 100

TEST RESULTS: (Compressive shear in psi values after 3 cycles)
ASTM D-905-49

	TESTED AT 75°F	TESTED AT 160	1°F
PRODUCT A		FAILURE 692 SIVE FAILURE	
PRODUCT B	1130 - 90% COHE 10% WOOD		

THE VALUES OF PRODUCT A COMPARE FAVORABLY WITH RESULTS OBTAINED FROM A STANDARD PHENOL RESORCINOL ADHESIVE USED IN THE INDUSTRY.

PRODUCT B SHOWS SLIGHTLY LOWER VALUES AT 160°F

The above test is required under the accelerated aging section of the ICBO Research Committee Acceptance Standard for Sandwich Panels Tentative Draft (January, 1974). This cycle is used for Class 2 Adhesives which see exterior exposure.

Table V compares the rate of strength buildup of both Product A and B versus various (competitive) established products (W to Z) in the marketplace.

This tabulation also shows the effect of 48 hour water soak on compressive shear values of Douglas fir/Douglas fir bonds assembled per ASTM D-905-49. Several common two part systems with limited shelf life (4 to 8 hours at 70°F or 1 hour at 90°F) are also included for comparisons.

In all cases, the cure rate of both Product A and B is considerably faster than any of the standard products listed. Resistance to water soak of both Product A and B is also much better than the majority of the room temperature curable products currently used in the marketplace.

TABLE V

# COMPARATIVE DATA TO EXISTING STANDARD PRODUCTS

FE SHEAF  905-49).  48 HRS WATEP  SOAK (PSI)  625  29  70-100  0	COMPRESSIVE SHEAR  (ASTM D-905-49)  AFTER 7 DAYS  a 77°F/50% RH 48 HRS  (PSI) SOAK 6  1230 625  1100 500  1100 500  1570 0  1350-1500 850-95	ULTIMATE STRENGTH (TIME) 1050 (24 HRS.) 1000 (16-24 HRS.) 500 ( 7 DAYS) 830 (24 HRS.) 1350 830 (3 DAYS) 870 (3 DAYS)	1-1/2 - 2 HRS.  3-4 DAYS  1-1/2 - 3-1/2 HRS.  2 DAYS  3 - 24 HRS.	TIME TO REACH  100 PSI  4'  30' - 1 HR.  6 HRS.  1 - 1-1/2 HRS.	PRODUCT (TYPE)  A (URETHANE)  B (URETHANE)  K (SYNTH RUBBER)  X (PVA)  Y (CASEIN 2 PT.)
29 70-100 0 850-950	294 1100-1750 1570 1350-1500	500 ( 7 DAYS) 830 (24 HRS.) 1350 830 (3 DAYS) 675 (1-4 DAYS)	3-4 DAYS 1-1/2 - 3-1/2 HRS. 2 DAYS 3 - 24 HRS.		(SYNTH RUBBER)  X (PVA)  Y (CASEIN 2 PT.)  Z (URFA FORMALIDEHYDE
58	787	500 ( 7 DAYS)		6 HRS,	W (SYNTH RUBBER)
200	1100	1000 (16-24 нкs.)		-	
972	1230	1050 (24 HRs.)	1	, †	A (URETHANE)
48 HRS WATER	a 77°F/50% RH (PSI)	ULTIMATE STRENGTH (TIME)	007	TIME TO REAC 100 PSI	PRODUCT (TYPE)
905-49).	COMPRESSIV (ASTM D- AFTER 7 DAYS				

WE ARE ONLY IN THE INITIAL STAGES OF AN OVERALL WOOD ADHESIVE DEVELOPMENT PROGRAM BASED ON URETHANE TECHNOLOGY.

THE PRECEDING DATA WAS PRESENTED TO ILLUSTRATE MOST OF WHAT WE KNOW TO DATE REGARDING THE CAPABILITIES OF TWO NEW PROTOTYPE ONE-PART ADHESIVES.

These adhesives cure fast at ambient temperatures, require little pressure applied to bonds during cure, and have good water resistance. One of them, Product B, has excellent gap-bridging properties (up to 1/16 inch), excellent flexibility (150% free-film elongation), and bonds well to damp wood.

FURTHER TESTING-WILL, OF COURSE, BE NECESSARY TO DETERMINE JUST WHAT KIND OF END USES THESE PRODUCTS (OR MODIFICATIONS OF THESE PRODUCTS) WILL BE SUITABLE FOR. THEY SHOULD WORK QUITE NICELY FOR MANY INTERIOR APPLICATIONS. THEY WILL PROBABLY BE QUITE SATISFACTORY FOR SOME EXTERIOR APPLICATIONS AND UNSATISFACTORY FOR OTHERS WHERE THE ULTIMATE IN LOAD BEARING PROPERTIES AND ENVIRONMENTAL RESISTANCE IS NEEDED; FOR EXAMPLE, IN GLULAM STRUCTURAL BEAMS. BOTH PRODUCTS ARE CURRENTLY BEING TESTED AGAINST THE STRINGENT SHEAR STRENGTH, DELAMINATION AND RESISTANCE TO CREEP REQUIREMENTS OF THE ASTM D-2559 SPECIFICATION ("ADHESIVES FOR STRUCTURAL LAMINATED WOOD PRODUCTS FOR USE UNDER EXTERIOR EXPOSURE CONDITIONS").

MUCH WORK MUST ALSO BE DONE IN DEVELOPING TECHNIQUES FOR APPLYING THESE FAST MOISTURE CURING, ONE-PART SYSTEMS IN AN EFFICIENT MANNER WITHOUT EXCESSIVE WASTE. NO DOUBT, VISCOSITY AND RHEOLOGY MODIFICATIONS WILL HAVE TO BE MADE WHERE LARGE SURFACE AREAS HAVE TO BE COVERED AND THIN BONDLINES ARE NECESSARY SINCE THESE CURRENT PRODUCTS CAN ONLY BE EXTRUDED OR CAULKED INTO A BONDLINE.

# NEW IMPROVED TWO-PART URETHANE ADHESIVE SYSTEMS

New improved two-part urethane formulations have recently been developed which perform better than our previously introduced SCOTCH-WELD® Brand Polymer Bond Structural Wood Adhesive Systems (i.e. 3555 B/A), particularly in the area of water resistance.

This development program is in the early stages and only limited data is available, but the data obtained to date is very encouraging. Several adhesive formulations are now under consideration and a sampling program will not be initiated until sometime after January 1, 1976. These adhesives cure quickly at room temperature with minimum pressure to form bonds that can exceed the strength of the materials joined.

# TABLE VI

# TYPICAL PHYSICAL PROPERTIES

BASE ACCELERATOR

COMPOSITION SYNTHETIC RESIN SYNTHETIC RESIN

COLOR YELLOW-AMBER DARK BROWN

VISCOSITY THIN LIQUID THIN LIQUID (150-300 cps)

MIX RATIO: BY VOLUME 1

WORK LIFE:

(25 GRAM QUANTITY a 75°F) 2 - 30 MINUTES

TIME TO REACH 80% OF

FULL CURE: APPROXIMATELY 10 - 120 MINUTES

THE MOST PROMISING FORMULATION TO DATE HAS BEEN TESTED FOR WATER RESISTANCE AND STRENGTH AT 160°F VERSUS A WIDELY ACCEPTED.

COMMERCIALLY AVAILABLE, TWO-PART PHENOL-RESORCINOL EXTERIOR GRADE ADHESIVE. THE RESULTS ARE TABULATED IN TABLES VII AND VIII.

AND TYPICAL PREVIOUS "POLYMER BOND" VALUES ARE LISTED FOR COMPARATIVE PURPOSES IN TABLE VIII.

# TABLE VII

# TEST RESULTS VERSUS AITC-110 CYCLIC DELAMINATION TEST

PROCEDURE: (Same as Table III)

RESULTS AFTER TWO CYCLES: (Less than 5% delamination required)

COMMERCIAL PASSES

PHENOL - RESORCINOL

NEW TWO-PART URETHANE PASSES

#### TABLE VIII

# RESISTANCE OF ADHESIVES TO CYCLIC LABORATORY AGING CONDITIONS

(ASTM D-1182-70 PROCEDURE C "Modified")

# CYCLE:

PERIOD IN HOURS	TEMPERATURE, °F	RELATIVE HUMIDITY, %
48	160 ± 5	10
48	73.4 <u>+</u> 2	IMMERSED IN WATER
8	-40	ABOUT 100
64	100 + 3.5	ABOUT 100

# TEST RESULTS:

# COMPRESSIVE SHEAR PER ASTM D-905-49 - NO AGING

	TESTED a 75°F	TESTED a 160°F
COMMERCIAL	1208 psi -	NOT TESTED
PHENOL-RESORCINOL	100% WOOD FAILURE	
NEW TWO PART URETHANE	1404 psi - 100% WOCD FAILURE	916 PSI - 90% WOOD FAILURE
OLD "POLYMER BOND" 3555 B/A	1725 PSI - 100% WOOD FAILURE	719 PSI -

# COMPRESSIVE SHEAR PER ASTM D-905-49 AFTER AGING 3 CYCLES

(PER ASTM D-1183-70 PROCEDURE C DESCRIBED ABOVE)

	TESTED a 75°F	TESTED a 160°F
COMMERCIAL	904 PSI -	533 PSI -
PHENOL-RESORCINOL	100% WOOD FAILURE	100% WOOD FAILURE
OLD "POLYMER BOND"	198 PSI -	
3555 B/A	NO WOOD FAILURE	
NEW TWO PART SYSTEM	1616 PSI - 75%	900 PSI - 60%
	WOOD FAILURE	WOOD FAILURE

FROM TABLE VIII, IT CAN CLEARLY BE SEEN THAT THE NEW FORMULATION HAS MUCH IMPROVED WATER RESISTANCE. IT ALSO DISPLAYS STRENGTH PROPERTIES AT LEAST EQUIVALENT TO THOSE OF THE PHENOL-RESORCINOL ADHESIVE.

ALTHOUGH THERE IS A WIDE SCATTER IN THE ABOVE TEST RESULTS, IT

CAN BE CLEARLY SEEN THAT THE STRENGTH OF THE NEW TWO-PART ADHESIVE

IS AT LEAST EQUIVALENT TO THE PHENOL-RESORCINOL ADHESIVES.

THIS TWO-PART STRUCTURAL MATERIAL OFFERS HIGHER COMPRESSIVE SHEAR VALUES THAN OUR ONE-PART SYSTEMS, BOTH INITIALLY AND AFTER ENVIRON-MENTAL AGING.

APPLICATION OF THESE FAST CURING TWO FART STRUCTURAL PRODUCTS REQUIRE
TWO-PART METER MIXING EQUIPMENT AND CLOSELY CONTROLLED HANDLING
TECHNIQUES WHICH HAVE BEEN PREVIOUSLY WORKED OUT FOR OUR "SCOTCH-WELD" BRAND POLYMER BOND ADHESIVE.

Additional testing is underway to evaluate performance under other specifications such as ASTM D-2559.

#### SUMMARY

BOTH ONE AND TWO-PART PROTOTYPE URETHANE ADHESIVE FORMULATIONS WHICH EXHIBIT FAST ROOM TEMPERATURE CURE RATES HAVE BEEN DEVELOPED. THESE MATERIALS FORM GOOD WOOD TO WOOD BONDS WITHOUT THE NEED FOR HIGH PRESSURE DURING THE BONDING STEPS. THESE CURED SYSTEMS DISPLAY WATER RESISTANCE WHICH IS SUPERIOR TO MOST OF THE COMPETITIVE SYSTEMS TESTED. ONE OF OUR PROTOTYPE ONE-PART ADHESIVES HAS EXCELLENT GAP FILLING CHARACTERISTICS OF OUR NEW TWO-PART SYSTEMS, WHILE THOUGHT TO BE GOOD, ARE JUST NOW BEING EVALUATED.

OUR ONE-PART SYSTEMS WILL LIKELY FIND USES WHERE SPEED OF CURE ENERGY CONSERVATION, BONDING SIMPLICITY AND GAP FILLING PROPERTIES ARE IMPORTANT. THE TWO-PART SYSTEM WILL LIKELY BE TRIED IN AREAS WHERE THE HIGHEST STRUCTURAL STRENGTH IS REQUIRED, BOTH BEFORE AND AFTER ENVIRONMENTAL AGING.

FURTHER FORMULATION WORK AND MORE EXHAUSTIVE TESTING WILL BE NECESSARY
IN BOTH THE ONE AND TWO-PART URETHANE ADHESIVE AREAS. New FASTER
CURING SYSTEMS WITH MODIFIED HANDLING CHARACTERISTICS, SUCH AS
SPRAYABLE OR FLOW COATABLE VISCOSITIES, WILL HAVE TO BE DEVELOPED
IN ORDER TO SATISFY MANY OF THE NEEDS OF THE VERY DIVERSE WOOD BONDING
INDUSTRY.

#### D. V. MADLE

#### ADHESIVE DEVELOPMENTS IN NEW ZEALAND

Although New Zealand is a small market by world standards, a wide range of adhesives for local industries is provided. The largest consumer of adhesive is the particleboard industry. Most of the particleboard produced is used for flooring and is high density material (43 lb/ft<sup>3</sup>) bonded with urea-formaldehyde resin at a relatively high loading. For this application, adhesives have been developed that ensure maximum durability with minimum evolution of formaldehyde fumes during and after manufacture. Development work has also been carried out on melamine-reinforced ureaformaldehyde resins, in order to increase the durability of the particleboard.

Studies have been undertaken to correlate the various laboratory methods for assessing durability, with exterior exposures.

Phenolic resins have been developed to supply a medium-density fiberboard plant, soon to come into production in New Zealand. Independent trials have shown the adhesives to be comparable, if not better than, similar resins currently used for fiberboard in other parts of the world.

A considerable amount of developmental work has been carried out on tannin-based adhesive systems for particleboard and plywood. Both South African mimosa tannin and locally produced radiata bark tannin have been tried. Satisfactory results have been achieved with particleboard in particular.

Due to the relatively small and fragmented nature of the plywood industry in New Zealand, developments with plywood adhesives have not progressed as rapidly, or as far, as those with particleboard adhesives.

#### ADHESIVE DEVELOPMENTS IN NEW ZEALAND

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Although New Zealand is a small market by world standards, the development of adhesives for the local wood-based industries has been quite intensive. In a small country like New Zealand, a big disadvantage encountered by an adhesive manufacturer is that many different types of glue are required. and in only relatively small quantities. However, this difficulty is partially overcome by producing multi-purpose adhesives, as opposed to tailor-made products. The production of multi-purpose adhesives also tends to give the consumer more durable products, since some properties are better than a particular end-user would demand. Nevertheless, New Zealand does have one large tonnage adhesive product (ureaformaldehyde resin for particleboard) and this enables a company like ICI to devote more time and money to developing adhesives for the smaller volume products.

Most adhesive is used in the particleboard industry and an indication of the amount involved can be gauged by New Zealand particleboard production figures relative to some other countries (see table II).

The per capita production figures (see table I) give a better indication of the importance of the particle-board industry in various countries.

TABLE I

PARTICLEBOARD PRODUCTION IN 1973 (AS 3" BOARD)

COUNTRY	M <sup>2</sup> PER CAPITA
New Zealand Sweden U.S.A. Australia Britain France W. Germany	1.79 5.43 1.57 1.35 0.30 1.99 4.73

(Source: ICI NZ 1975)

Significant developments have occurred in New Zealand with tannin adhesive systems, low fume urea-formaldabyde resins and medium density fibreboard resins. Before discussing these in more detail, however, it would be helpful to briefly outline the development of the particleboard industry in New Zealand, since most adhesive developments followed on from this.

#### History

In 1958, the first New Zealand-made particleboard was produced by the Fletcher Timber Company (FTC) in Christchurch and used on a trial basis in housing construction. Pavourable results from these trials led to the usually conservative, government homefinance institution approving particleboard use as a flooring material. Due to certain advantages over conventional strip flooring, the demand for flooring grade particleboard rapidly increased throughout the 1960's, thus causing a corresponding increase in the output of urea-formaldehyde (UF) syrup. New particleboard plants were opened in 1965 (Auckland), 1969 (Taupo) and 1973 (Kumeu, near Auckland) and, to meet demand, new adhesive producing plants in 1970 (Mt Maunganui) and 1972 (Christchurch). The present position is that over 75% of new houses and flats are now built with particleboard flooring compared to only 15% with other wood products, such as tongue and groove flooring, and less than 10% concrete.

In contrast to particleboard, plywood production has not changed greatly over the past ten years and prior to 1964, plywood adhesives were imported. Although acceptable resins can be supplied, the impetus for improved adhesives can only come with the introduction of more sophisticated plywood wills in New Zealand.

The laminated timber industry has grown steadily over the past seven years. The use of such timber for structural applications such as beams is now widespread.

Fingerjointing of Pinus Radiata lumber to remove knots and upgrade it, started in 1967. The product is used mainly for siding (weatherboard) and utilises a resorcinol resin developed by ICI.

### Particleboard Adhesives

The particleboard industry is the largest consumer of adhesives; almost all of which are urca-formaldehyde types. Figures for particleboard production are given in table II.

TABLE II
PARTICLEBOARD PRODUCTION

Country	Year	Production $(m^3)$	Particleboard mills
New Zealand	1974	118,767	5
Sweden	1973	842,000	14
U.S.A.	1973	6,283,000	72
Australia	1973	338,000	10

(Source: World Wood 1975)

The two companies producing particleboard in New Zealand (FTC and New Zealand Particle Roard Atd) use a UF syrup of about 65% solids. About 80% of production compaises a 20mm (approximately %"), high density (431b/in<sup>3</sup>) flooring grade board that contains wax.

Unlike in many countries, this board is permitted for flooring use without underlay, provided there is good underfloor ventilation. So far, this has been found satisfactory if relatively high levels of resin (10% and above), together with good quality wood chips and a suitable amount of wax are used in board manufacture. Undoubtedly a big factor in the official acceptance of particleboard flooring has been the equable climate of New Zealand. Both average temperature and humidity are such, that there is no adverse effect on board durability if proper precautions are taken. Some climatic comparisons between the United States and New Zealand are shown in table III.

Until now, the major user problem has been formaldehyde fumes released from the fresh particleboard. To minimise this, ICI has studied methods that reduce fumes without impairing the durability of the finished board. With UF resins, it is possible to reduce the free formaldehyde fumes by about half, without any loss in durability, by careful optimisation of resin and hardener (this is the U242-DD hardener system shown in table TV). The effect of this system can be compared to results for an ordinary particleboard resin (ICI U240) with an ammonium chloride - hexamine hardener (using, per 100g of uncatalysed resin, in the surface 0.128g ammonium chloride with 0.217g hexamine and in the core 0.297g ammonium chloride with 0.127g hexamine).

This latter system has been used as a typical particle-board adhesive in New Zealand for several years. The results given for boards D and E are indicative of the best results obtainable with the system.

A comparison between boards A, B and C and between boards D and E, shows the effect of decreasing the mat moisture. With the ICI resins, this lowers the free formaldehyde level in the board, increases the amount of swelling and increases durability as measured by MOR and IB retention after 6 cycles of ASTM D1101-59 (TB retention being 46%, 54% and 56% respectively for boards A,B, and C).

The degree of reduction in free formaldehyde level is shown by comparing the effect of the DD hardeners (learneds A,B, and C) with the AA hardeners (boards D and E). Despite a marked reduction in fumes, results from these and other board tests show no significant decrease in durability on changing from the U240-AA hardener system to the U242-DD hardener system.

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LETVER SOME CITIES IN NEW ZEALAND AND THE UNITED STATES.

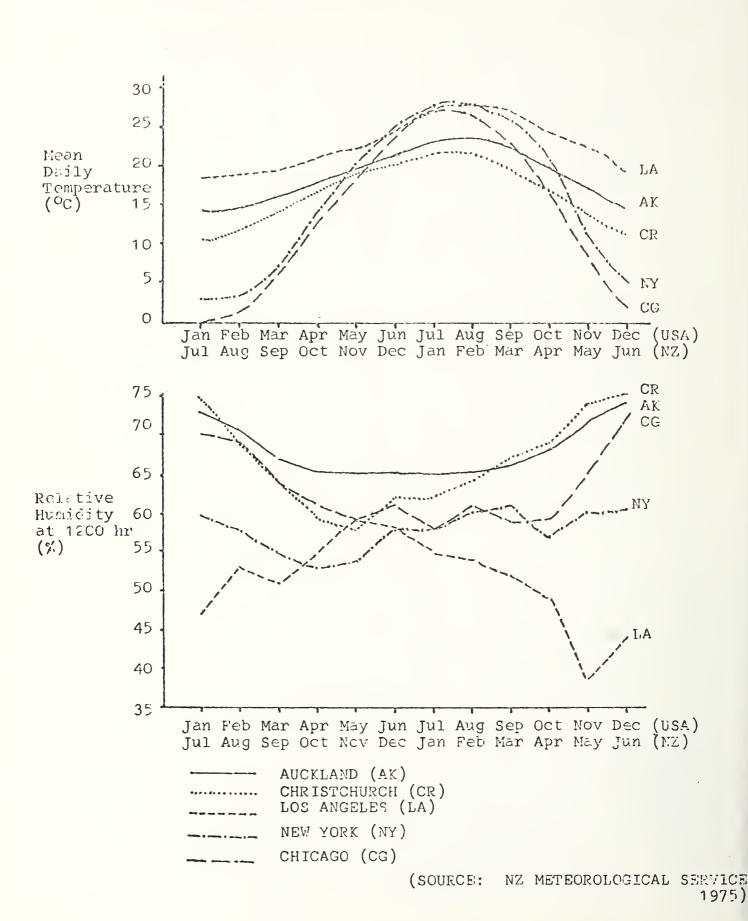


TABLE IV

EFFECT OF DIFFERENT UF REGIN AND HARDENER SYSTEMS ON FREE FORMALDEHYDE AND DURABILITY OF PARTICLEBOARD

Formaldenyde	Age of board (days)	12	30	7	30	10	27	v	Testing and priate 31-59.
Free Form	Value (x 10 <sup>-</sup> 3%)	26.5	24.2	. 14.7	35.2	28.8	18.0	35.4	for Testing Materials S D1101-59.
01	Spring- t back (%)	.0 6.5	٠. و ن	24.4 13.8	.6 4.5	.3 10.9	.8 10.9	0.	
ASTM 110	$\frac{NOR}{b/in^2}$ $\frac{1'I}{Wet}$	2465 16	2820 17	3340 24	3450 14	3730 19	1602 23	3544 23	Moisture in core rnal Bond r Absorption WA=O% American Society
6 cycles /	1B M(1b/1b)	70 2	99	87 3.	70 3.	136	37 10	33	Mat Moisture in c Internal Bond Water Absorption å so WA=C% 1101.American Soc
1811)	hour cold er soak	16.1	15.2	28.5	17.2	4.61	15.9	23.0	C: IB: WA: Grie
per BS	24 hour water s	7.7	0.5	14.8	8.7	11.3	8	12.0	est piece
(Tested as	(1b/in?)	153	121	155	146	506	100	151	ace   when test   1811:1961
	MOR 15/in9	4943	4906	4295	4410	4421	3092	3523	
Properties	Moist-MOR ture (%)(1b/in <sup>2</sup> )	11.0	9.7	7.2	11.7	4.	12.0	9.7	Mat Moisture in sur Modulus of Rupture Thickness increase Igback is based on 7
3" Board	Density (1b/ft3)	44.3	46.5	45.7	45.6	44.8	48.3	44.7	S: Mat Moisture in surf MOR: Modulus of Rupture TI: Thickness increase Springback is based on TI BS 1811 British Standard
Average Mat Mois-		19.8 (s 23.9/c 15.7)	15.0 (s 17.6/c 12.4)	11.1 (s 12.6/c 9.6)	19.8 (S 23.9/c 15.7)	11.1 (s 12.6/c 9.6)	19.8 (s 23.9/c 15.7)	11.1 (S 12.6/c 9.6)	Used: S: MOR TI: Spr BS
	System	U242 DD hardener	U242 DD	U242 DD	U240 AA hardener	U240 /A	American 19.8 (self-cat-23.9/calysed UF15.7)	American 11.1 (S self-cat-12.6/c alysed UF9.6) resin	Abbreviations
	Board	⋖	m	0	U	ជា	(I.	U	Αὸρινε

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The critical effect that hardener choice can have is further shown by comparisons between boards with the ICI hardener system (where the free formaldehyde level is less at the lower mat moisture) and those (boards F and G) with an American self-catalysed resin (where the free formaldehyde level is actually less at the higher mat moisture).

Free formaldehyde was determined by the method of R Christensen (ref: Forest Products Journal 1972 22(4) 17). The age of the tested board, and the temperature and storage conditions, affect the free formaldehyde so results shown cannot be regarded as absolute values, but merely as an indication of trends. In fact, for a given board, the free formaldehyde result is dependent on the part of the board that is tested. Free formaldehyde results varying between 10 x 10-3% and 22 x 10-3%, for example for the same board are not untypical.

It is essential therefore that care be taken when sampling for these tests, and a standard procedure needs to be used, if meaningful results are to be obtained.

It is also appropriate at this stage, to discuss the methods used for assessing durability. This is measured by subjecting the board to six cycles of the ASTM D1101-59, which involves a vacuum-pressure cold water soak cycle, with drying at 300-350C. After six cycles, the thickness increase is measured wet (TI wet) and then the test piece is dried to the original (starting) moisture level. This gives a "springback" figure. The internal bond (IB) and modulus of rupture (MOR) is also measured at this stage.

It has been found that this procedure gives 50-70% strength retention for "good" urea-formaldehyde bonded boards. By comparison, boards that contain melamine reinforced UF adhesive (10-30% melamine solids) give 90-100% strength retention. Poor UF boards can delaminate in this procedure despite good initial strength values.

Although no problems have been encountered so far, particleboard has only been used in flooring situations in New Zealand for 10 years. This compares with the average life of a house, which is in excess of 50 years. Should defects arise in a house during the term of a mortgage, then the leading institutions in New Zealand could reconsider their approval of UF bonded particle-board flooring. Such a move would naturally have far reaching effects on adhesive suppliers and particle-board manufacturers. It is for this reason that durability is the prime concern in the development of particleboard adhesives.

Improved durability of particleboard has been achieved by using melamine reinforced UF resins. Despite the extra cost of these modified adhesives, a lot of work has been carried out in this area. However, to date, no melamine reinforced board is being marketed in New Zealand. Typical board test results are shown in table V for comparison purposes. The 6 cycle ASTM D1101-59 mentioned earlier is not severe enough to show durability defects. Results of the 2 hour boil test or the French Centre Technique du Bois Standard V313 test are therefore preferred.

 $\frac{\text{TABLE V}}{\text{Comparison of durability test results}}$ 

Boil	IB (% retention	nates	41	65	ο <i>)</i> ∞
2 hour Boil	Springback (%)	delaminates	14.8	5.	7.6
3	IB (% retention)	4	85	65	ı
V313	Springback (%)	23.8	e. 9	2.7	ſ
101	MOR (% retent- ion)	41	100	∞ 4.e	100
cles ASTM 1101	IB (% retent- ion)	46	6	97	100
6 cycles	Spring-	21.5	9.	2.7	3.0
Density	(1b/ft³)	47.1	42.5	43.8	43.0
ر به در ۱۲۰۰ در ۱۳۰۰ در ۱	rarticienoara	Typical NZ commercial UF board (3" flooring grade)	30% melamine/UF $(\frac{3}{4}$ ") ICI	French com- mercial mela- mine/UF with additive (0.712)	Mimosa tan- nin $(\frac{3}{4}$ ") ICI
90	<b>9</b> 000	Ĺt,	M25	ъ К	ਸ74

(SOURCE: ICI NZ 1975)

#### Tannin Adhesives

In New Zealand, much interest has been shown in the development of tannin (Mimosa and Radiata) adhesive systems for exterior grade particleboard and plywood. Work has been carried out by the particleboard manufacturers, and ICI, to develop competitively priced durable products.

The advantages of a Mimosa tannin particleboard are: its low cold water swelling characteristics; its price when compared to boards utilising adhesive made from high cost imported phenol; its need for a short press time that is more comparable to UF than phenolic resins. One New Zealand company (FTC) is marketing a particleboard made with an imported Mimosa tannin adhesive and ICI had developed a similar system using imported Mimosa bark extract.

Since Mimosa tannin has to be imported from South Africa, development work has proceeded to produce an adhesive based on locally produced Radiata tannin. A pilot plant has been set up in New Zealand by one company (NZ Forest Products Limited) to extract the tannin from the plentiful supplies of Pinus radiata bark. ICI has produced experimental particleboards that use a Radiata tannin adhesive (test results for a typical board, and a board using the ICI Mimosa tannin adhesive, are shown in table VI).

Tannin particleboards are comparable in durability to phenolic resin bonded boards, although a higher resin loading is necessary. However in outside exposure trials, the surface layers show more of a tendency to deteriorate, when compared to phenolic boards, if the board is left unpainted.

TABLE VI

Test results for experimental tannin boards

do	Tannin	Board	Prop	erties	s (Tes	sted as per	BS1811)		2 hour Bo	oil
	Adnesive	Thick ness (in)	Den- sity (1b/ ft <sup>3</sup> )	MOR (1b/ in <sup>2</sup> )	IB (lb/ in <sup>2</sup> )	24 hr cold Ti (%)	water soak WA (%)	TI Wet (%)	IB (lb/in <sup>2</sup> )	Spring- back (%
7	Mimosa Raciata	0.75	48.0 48.4	3730 3670	110 121	6.9 8.5	11.9 15.2	15.2 17.9	108 96	7.6 7.2

(SOURCE: ICI NZ 1975)

#### Medium Density Fibrebourd

Medium density fibreboard (41 lb/ft<sup>3</sup>) is soon to be produced by a plant in Christchurch. Total production capacity will be of the order of 110 tonnes per day.

Phenolic resin bonded fibreboard for exterior siding, and UF bonded fibreboard for furniture and flooring, will be produced. Resins have been developed by ICI over the last twelve months for use in these boards, and during independent trials in Sweden, they were found to give results as good as, if not better than, results from American fibreboard resins. Table VII shows the effect of different phenolic resins on fibreboard properties.

TABLE VII

Properties of some experimental fibreboards
(11mm thickness)

Standard press conditions - tested as per Swedish standard \$15 23 2901

Phenotic	Resin Loading	Board Density	MOR (1b/in <sup>2</sup> )	IB (1b/in <sup>2</sup> )	2 hr cold w	ater soak
Resin	on dry fibre (%)	(lb/ft <sup>3</sup> )	(15) 111 )	(10/111)	ті (%)	WA (%)
ICI Resin 113	7.2	43	6480	68	3.3	11.2
ICI Resin 133	6.6	43	7250	82	3.0	9.6
US Fibre- board Resin	7.1	43	6750	78	4.8	14.7
Australian Resin	7.1	43.5	5630	60	22.0	56.0
Swedish Resin	6.7	43.5	5280	33	6.1	12.8

(SOURCE: DEFIBRATOR AB 1975)

#### Plywood

The New Zealand plywood industry is relatively small and fragmented. Total output is low at present, due to a hortage of suitable logs. Recent production figures are shown in Table VIII.

TABLE VIII

#### Plywood production (hardwood and softwood types)

Country	Year	Production (m <sup>3</sup> )	Plywood mills
New Zealand	1974	105,000	5
Sweden	1973		7
U.S.A.	1973		396

(SOURCE: WORLD WOOD 1975)

Almost all plywood in New Zealand is made from softwood and Pinus radiata is the dominant species (60% of peeler logs). The market for plywood is such, that a new mill for softwood ply is soon to go on stream (at Kinleith) and this will effectively double present capacity.

Satisfactory urea-formaldehyde and phenolic resins have been developed for use as plywood adhesives.

#### Other Areas

Most fingerjointing is done by continuous high speed RF curing of resorcinol, melamine and urea resins. However some firms still cold cure. Resins of all these types have been developed by ICI in New Zealand, and improvements in user handling characteristics and economy are continually under investigation. Before the resorcinol shortage only resorcinol-phenol adhesives were permitted for use in fingerjointed siding (weatherboard), but now melamine-UF resins have been approved. A stable liquid melamine-UF resin has been developed by ICI.

Hot melt adhesives are tending to replace separate application UF adhesives for clashing strips on doors.

New Zealand has a continuous belt process (Bison-Mende) for thin particleboard, which utilises urea formaldehyde resin. The firm () making this board experienced early marketing and quality control problems. There is still some consumer resistance to the product, although this has now been overcome by a much improved board.

#### Conclusion

Most adhesive usage is in products for home consumption and, at present, little is exported. The ready acceptance by the New Zealand public of the adhesive containing products has ensured the growth of the industry. The emphasis on durability hopefully means that the products will maintain their established position in the construction industry.

# APPENDIX Location of Major Adhesive Using Plants in New Zealand (1975)Kumeu-Auckland • Major Centres of Population. ▲ Particleboard Mills. • Plywood Mills. Mt Maunganui ■ Medium Density Fibreboard Plant, Hamilton \* Adhesive Manufacturing Plants. Main Exotic Forests (Indigenous Forest Nct Shown). Kimleith (Blag Taupo Scale: 100 miles to 1 inch 0 Lower Hutt Wellington Greymouth Christchurch 23% of land area is forest. Dunedin 5.8% of land area (4 million acres) is considered to be merchantable forest.

#### JOHN D. ROSE

John is head of the Construction Applications Section in the Applied Research Department of the American Plywood Association. He is responsible for developing and conducting structural tests on new and improved plywood construction systems for homes, apartments, light commercial buildings, and manufactured housing. He graduated from the University of Michigan with a BS degree in Structural Civil Engineering, and has been with the American Plywood Association since 1966.

#### USE OF ADHESIVES IN CONSTRUCTION - WHAT'S NEXT?

Construction adhesives have gained widespread usage in the past 10 years for field-gluing plywood floors to joists. Adhesives in this application improve the performance of the floor and offer tangible benefits to both the builder and homeowner. The successful experience with construction adhesives has led many builders and adhesive manufacturers to inquire about other potential uses for field-applied adhesives in construction, such as installation of wall sheathing and siding, roof sheathing, and fabrication of plywood components. The advantages and disadvantages of using adhesives in these applications are discussed, and other factors influencing their use are reviewed. Current research and development projects involving use of construction adhesives are highlighted, and needs for technical information on adhesive properties and structural performance of completed assemblies are outlined.

# USE OF ADHESIVES IN CONSTRUCTION - WHAT'S NEXT?

John D. Rose American Plywood Association

September 25, 1975

Prepared for presentation at the 1975 Symposium
"Adhesives for Products from Wood," sponsored by USDA Forest Service, Forest Products Laboratory, Madison, Wisconsin

#### SUMMARY

Construction adhesives have gained widespread usage in the past 10 years for field-gluing plywood floors to joists. Adhesives in this application improve the performance of the floor and offer tangible benefits to both the builder and homeowner. The successful experience with construction adhesives has led many builders and adhesive manufacturers to inquire about other potential uses for field-applied adhesives in construction, such as installation of wall sheathing and siding, roof sheathing, and fabrication of plywood components. The advantages and disadvantages of using adhesives in these applications are discussed and other factors influencing their use are reviewed. Current research and development projects involving use of construction adhesives are highlighted and needs for technical information on adhesive properties and structural performance of completed assemblies are outlined.

#### INTRODUCTION

When APA was originally approached on giving a presentation at this symposium, it was suggested that a discussion of field-glued floors might be appropriate. However, glued floors are well-known to many of you, so I'd like to only briefly discuss some aspects of research and development on this application and then concentrate on other plywood uses where construction adhesives appear promising. As in the case of glued floors, the usage of adhesives in other applications will depend on the advantages or disadvantages in structural or in-service performance, and the added cost, if any, of using adhesives.

#### FLOOR APPLICATIONS

Let's start first by looking at glued floors. Certainly, in the past five years, this application has proven to be a big market for both construction adhesives and plywood. For example, APA and NAHB studies of new construction in 1973 and 1974 show that about 25% of single-family homes and 20% of multi-family units were built with glued floors. Based on 1974 housing production of about 890,000 single-family units and 450,000 multi-family units, it's estimated that about 1.3 million gallons of construction adhesive were consumed for glued floors -- in a year of low housing production. Since active promotion of the APA Glued Floor System started only five years ago, after code acceptances were obtained, its widespread use in such a short period of time is truly amazing -- but not unexpected, since its popularity among builders can be traced to positive improvements in floor performance, while reducing construction costs.

In APA field calls and surveys, builders cited the reduction or elimination of floor squeaks and increased floor stiffness as the primary reasons for using glued floors. Both floor performance and quality of construction are improved at little added expense to the builder or home-owner. APA cost studies show that even if no other changes to the floor construction are made, use of adhesive to glue the plywood floor to the joists would add less than \$100 to the cost of a typical 1600 square foot home.

On the other hand, since the floor stiffness is increased, joists can be used on longer spans, or joist size reduced or spacing increased, while maintaining floor stiffness required by codes. In these cases, the added costs for applying adhesives and application are offset by reducing materials and labor costs, thereby providing additional advantages to the builder. In fact, it's hard to conceive of any disadvantages for glued floors.

Despite the fact that glued floors have been accepted by all building codes and have been actively promoted by APA for several years, research on the system by other organizations continues to provide new information on adhesive properties and structural design of glued floors.

In 1971, research by Kuenzi and Wilkinson at FPL provided a more theoretical method for calculating the stiffness of composite beams where non-rigid adhesives or fasteners were used (1). These important analytical methods have paved the way for research by other individuals to develop the design method more completely. In particular, Goodman and others at Colorado State University (2) and Hoyle at Washington State University (3) have identified factors affecting the stiffness and strength of beams and floors. These include the adhesive shear modulus (or stress-strain relationship); the effects of discontinuities at joints between plywood panels; and the load-slip relationship for combinations of adhesives and mechanical fasteners. Design formulas were developed to account for these variables. While the refined design methods used in recent research are admittedly more sophisticated than the method developed from APA tests (4), we have observed that practically the same end result will be obtained by these methods when the effect of actual construction conditions are taken into account.

One might ask whether there is still a need for further research or development on glued floors. The answer is yes.

First, a standard test method needs to be developed for determining the adhesive shear modulus -- in fact, it was pointed out yesterday that a test method is already being drafted by members of the ASTM D14.7 Subcommittee. Development of this information is basic to the design method used by Kuenzi, Goodman and Hoyle.

Hoyle points out that use of adhesives with shear moduli of 100 to 200 psi will produce assemblies almost as stiff as if rigidly glued (3). Even adhesives with low shear moduli (from 25 to 100 psi) can materially improve composite action.

The shear modulus depends on the stress level and the adhesive deformation per unit of glueline thickness. Based on a review of APA and ASTM test data and discussions with Hoyle, it was decided that an average glueline thickness of about .020" - .030" could be assumed for most glued floors.

Second, a probable reduction in joist bending stress, due to composite action between the plywood floor and joists, needs to be substantiated by tests and analysis. The amount of stress reduction depends on several factors, including the joist size and spacing, and whether plywood joints are glued or unglued. Tests should investigate how creep under long-term loads affects the stress level, and whether discontinuities at plywood joints cause stress concentrations at these locations. Hoyle points out that adhesive creep has only a minor effect on the creep of glued assemblies (3).

Third, the durability of adhesives used structurally -- for example, to reduce joist bending stress -- should be studied under long-term stress.

Standard test methods need to be developed, and could pave the way towards using construction adhesives in structural applications such as beams or trusses.

Simplified design methods are needed so that engineers and architects can design glued assemblies without requiring a complex analysis. Theoretical design methods can account for loading, geometry and various support or boundary conditions, but these conditions vary from floor to floor ---in addition to the basic variability of the materials used. Therefore, a "finely-tuned" design method is not warranted for general floor applications.

#### WALL APPLICATIONS

Because of the successful experience with construction adhesives in glued floors, we have received many requests from industrialized housing and adhesive manufacturers, inquiring about gluing plywood to wall studs. Research on this application has been undertaken by Atherton and Polensek at Oregon State University, Krueger and others at Michigan Technological University, and to a limited extent by APA.

First, let's review some of the advantages and disadvantages in gluing sheathing or siding to studs. It's generally recognized that gluing panels to the studs could increase their stiffness and strength under lateral, axial and racking loads. High racking or shear forces are sometimes encountered where short wall sections are required to resist wind or earthquake conditions. For example, the end walls in a plywood-sheathed warehouse in Tacoma are designed for shear loads over 1100 pounds/ft. To resist these high loads, close nail spacing or multiple layers of sheathing and siding are necessary. In such cases, glued shear walls could save labor and material costs by reducing nailing or eliminating extra sheathing. These special cases are not encountered very often, however.

A design method for glued shear walls was recently developed by Krueger (5).

Several years ago, APA tested 12 glued shear walls under dry and wet conditions.

The plywood in these tests was installed with a construction adhesive and 40% fewer nails than normally used. The ultimate shear loads for these walls was from 5 to 10 times the allowable design shear load for comparable, conventionally-nailed shear walls (ordinarily, load factors of 3 or 4 are anticipated). It was found that framing tolerances and nailing techniques can influence the strength of the shear wall, however.

Composite action with glued sheathing and studs can stiffen and strengthen the wall under lateral and axial loads. A corresponding reduction in the lumber grade is possible, or - more likely - higher lateral or axial loads could be permitted for existing stud grades. Research by Atherton and Polensek is underway to investigate the composite action of glued wall sections and to develop a design method for nailed and glued walls. In an extension of this application, APA has received a number of inquiries on gluing plywood sheathing to stiffen and strengthen studs which are oriented flatwise, to minimize the thickness of mating walls along the centerline of sectional or mobile homes.

Glued girder walls can stiffen and strengthen the "open side" of mobile and sectional homes during transportation. Such walls generally consist of sheathing or interior paneling glued to the studs to reduce or eliminate sticking doors and "cosmetic" damage at corners or joints on interior wall surfaces. APA has tested several 28-ft long girder walls with plywood exterior siding and drywall interior surfaces (6).

The walls were tested as simple span and cantilever beams under simulated transportation loads up to 2½ g, based on the estimated dead weight of a 12' x 28' unit. Surprisingly, the midspan deflection of the nailed walls was only about

1/16" greater than for a comparable glued wall. The basic stiffness of the 8-ft high wall -- even when nailed only -- seems to be adequate for this application. For mobile homes, however, gluing of light, thin interior paneling on only one side of the wall may stiffen the wall to a greater extent than observed in our tests.

Except for these special cases, glued walls do not appear to offer any particular advantages for conventional construction. Most nailed-only walls have adequate stiffness and strength to easily resist lateral, axial and racking loads.

Are there any specific disadvantages or problems which can result from gluing panels to wall studs? Gillespie noted yesterday that thin, lightweight wall sections tend to bow under differential moisture conditions. As an extension of this problem, APA is concerned whether adhesives restrict panel expansion under high moisture conditions, leading to panel buckling between studs. This is primarily of concern for siding, which is directly exposed to the weather.

Panels installed vertically, with the face grain parallel to stude, can aggravate the problem since plywood has lower stiffness across the panel width to resist buckling forces. An important fact to remember is that glued siding is difficult, if not impossible, to replace if buckling occurs.

Since the builder and home-owner demand superior appearance and performance for siding, it seems risky to suggest glued walls until the potential problems can be resolved. For the present, APA does not recommend gluing siding to studs. However, tests are underway to investigate siding buckling and to develop siding recommendations to insure satisfactory performance under all conditions.

It appears that buckling can be reduced or eliminated by using thicker panels -such as 5/8" plywood -- and for grooved siding, by using panels with widely-spaced
grooves, such as 12" o.c. Also, panels installed horizontally, with the face
grain across studs, have greater stiffness to resist buckling forces. Conventional double-wall construction, consisting of plywood sheathing glued to the
studs, and the siding installed with normal nailing, also appears promising. The
problem is just as likely to occur with other siding products, too.

Another potential disadvantage is the in-place cost of glued walls. For walls with study 16" o.c., 40 lf of adhesive bead is required, corresponding to an adhesive cost of about 75¢ per panel. The labor cost for applying the adhesive is estimated at about 45¢ per panel. By reducing nail spacing to 12" o.c. along panel edges and study, 24 nails can be eliminated, corresponding to a labor saving of about 30¢ per panel. The net additional cost of using adhesives is therefore about 90¢ per panel. The cost will be even higher if thicker (more expensive) siding is required to eliminate potential buckling problems.

Accordingly, it is felt that builders will not be interested in glued walls, except in cases where the wall must resist high shear loads. The "break-even" point appears to be when nails must be spaced about 2"-3" o.c. around panel edges.

Another point to be considered is that code recognition of structural values for glued walls will likely require that walls will have to be fabricated under the supervision of a special inspector approved by the local building department. These inspection conditions are already imposed by ICBO for glued lumber roof diaphragms <sup>(7)</sup>. They mean added costs, and more complicated construction processes for the builder.

There are a number of questions that must be answered before seeking code acceptance of structural glued walls. First, the adhesive shear modulus and strength should be determined, especially at elevated temperatures up to  $130^{\circ}$ F, which can occur inside walls according to a past FPL study<sup>(8)</sup>. Development of the test methods previously mentioned should aid in making this information available.

Second, structural tests under both dry and wet conditions should be performed to develop or substantiate design methods for glued shear walls; for stiffening and strengthening of studs under lateral and axial loads; and for girder walls. The tests should include evaluation of various materials which can be used for siding, sheathing, interior paneling and studs. This could be a monumental task, and it's complicated by the lack of reliable design data on many such products.

Third, exterior durability of construction adhesives should be evaluated, since the adhesives can be exposed to weathering at siding joints. This requires development of suitable durability test methods. Perhaps the exterior glue durability tests for plywood could be considered as a basis for this determination.

Fourth, recommendations need to be developed for installing glued siding to insure satisfactory performance and avoid potential buckling problems. As noted previously, this applies to all siding products which could be glued to studs.

Last, an economic feasibility study is recommended to define the conditions under which glued walls are practical. Really, this question should be answered first.

#### ROOF APPLICATIONS

Finally, let's turn our attention to roof applications. Glued roof diaphragms have been tested by Washington State University and Oregon State University, and structural values have been accepted by ICBO<sup>(7)</sup>.

For those of you who aren't familiar with this terminology, a roof diaphragm can be thought of as an I-beam laid on its side. Lateral loads from winds or earthquakes are resisted by the framing around the perimeter of the building, which is designed as a continuous "flange" member to resist bending forces, and by the plywood sheathing which acts as the beam "web" to resist shear forces.

In the Oregon State University tests on glued lumber diaphragms, the T&G edges of the lumber decking were glued with a construction adhesive (9). Tests showed that the adhesive substantially improved the stiffness and strength of the assembly, even when openings were cut into the deck for skylights or access doors. A side benefit is that composite action of the glued roof diaphragm will permit framing members to carry higher roof loads, just as in the case of glued floors.

Increased ICBO seismic loading requirements in California have prompted APA to take another look at ways to increase allowable design shear loads for plywood diaphragms. As in the case of high load shear walls, glued roof diaphragms offer potential cost savings by reducing nailing or eliminating the need for a two-layer plywood roof or thicker plywood. Tests are planned at APA to evaluate performance of glued plywood diaphragms and to develop design recommendations for high-load applications.

Many of the points relating to glued walls also apply to glued roof diaphragms. These include development of test methods to determine adhesive properties at elevated temperatures up to  $160^{\circ}$ F, which can occur in enclosed roof areas according to past FPL studies<sup>(8)</sup>; structural tests of glued diaphragms to establish design values and develop design methods; and economic feasibility studies.

In addition, since the glued roof diaphragm acts in a structural capacity, its construction will undoubtedly require the supervision of a special inspector as previously noted.

The potential advantages of glued roof diaphragms are limited to cases where the construction must resist high shear loads. In most cases, conventional nailed roof construction for residential and commercial buildings is adequate and use of construction adhesives is therefore unnecessary.

#### GLUED COMPONENTS

Because of the limited time, I have not attempted to cover construction adhesives for fabricating components such as beams and trusses. With the exception of mobile homes, present market requirements for components are satisfied by metal-gussetted trusses or manufactured beams such as glulams, Trus-Joist, Wood Bar or other similar components. Although research on glued components fabricated with construction adhesives has been conducted by Hoyle, Kuenzi and others, present market requirements for such components appear to be limited primarily to mobile and sectional homes.

In closing, I'd be happy to discuss any of these thoughts further with you, or attempt to answer any questions you may have.

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#### FORECAST

# OF THE FUTURE

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### JAMES A. KOUTSKY

Jim is Associate Professor of Chemical Engineering at the University of Wisconsin at Madison. He received his education at Case-Western Reserve, getting his BS and MS in chemical engineering and his doctorate in polymer science. His Ph. D. research was related to nucleation of semi-crystalline polymers and other low-molecular-weight liquids. His present work is focused on structural investigations of various solid polymers, with primary emphasis on thermosets and adhesives.

## FUNDAMENTAL RESEARCH ON ADHESIVES

Long-term stability of adhesives has been of primary concern for many years. In the area of structural adhesives, recent research has focused upon: (1) characterization of the mechanics of adhesive joints, (2) solid-state structure studies of adhesives, and (3) chemical stability of the adhesive joint to hostile environments.

One great difficulty in predicting performance of an adhesive system for a particular application is relating the fundamental mechanical property values of the adhesive (modulus, ultimate strength, elongation, fatigue resistance, etc.) to the complicated stress fields associated in composite joints. Application of an external stress to a test joint can generate internal stress fields which are considerably larger than those simply calculated from external loading. A very useful development has been the calculation of stress coefficients for different joint geometries.

Studies on the solid-state morphology of thermosetting adhesives have been initiated mainly to better understand the relationships of adhesive formulation and ultimate strengths of adhesives. Improvement of these properties is apparently related to the gel structure near interfaces. Normally, stress fields are highest at these interfaces and the long-term stability of a joint will depend upon the adhesive structure near the interface.

The chemical stability of adhesive resins has been previously correlated with the bulk polymer chemical structure. However, interfaces can play an enormous role in determining whether a joint can survive in a hostile environment. Newer correlations of relating thermodynamic wettability and stability of adhesive systems have been attempted. Recent durability studies have also shown the value of using thermodynamic wettability criteria to calculate the long-term stability of adhesive joints to water exposures. Also, chemical reactions of an adhesive to a substrate have been modeled as time relationships with the fraction of bonds formed. This model has simply correlated mechanical performance of the adhesive joint and various chemical parameters.

#### THOMAS H. ELLIS

Tom is Project Leader for Marketing Analysis and Economics of Forest Products Utilization at the Forest Products Laboratory. He received his B.S. in Forestry from the University of the South, his M.F. in Mensuration from Yale, and completed the course work for Ph. D. in Forest Economics at Michigan State. Before joining the FPL staff, he was Timber Program Analyst in the Division of Programs and Policy Analysis for the Forest Service in Washington, D.C.

#### FUTURE EXPECTATIONS FOR BONDED WOOD PRODUCTS

The most recent projections of timber products demand and supply made by the U.S. Forest Service suggests that there will be substantial increases in demand for plywood and particleboard in the next 5-15 years. As compared to 1973 consumption, softwood and hardwood plywood demand is expected to increase 40 to 50 percent and particleboard, 65 percent. These projections are based principally on assumptions that gross national product and disposable personal income will rise at rates of 3-1/2 to 4 percent per year in constant dollar terms; that rates of technological change in forest industries will continue about the same as experienced in the past 10-15 years; and that prices of timber products, relative to prices of all commodities and to most competing materials, will not change significantly from the 1970 levels and those levels experienced in 1975.

There are great uncertainties in these underlying assumptions. It is not yet clear how rising energy costs will affect the competitiveness of wood products nor how the U.S. economy will be affected in general by rising energy costs. Eventualities such as a shift to construction of smaller houses or a shift in the type of housing demand could result in considerably lower demand for wood products than indicated above.

Glue costs obviously are a major factor determining the competitiveness of bonded wood products. Availability and price of wood furnish is another major factor of uncertainty. If sawmill efficiency increases as expected, and sawlog consumption is no greater than anticipated, supplies of mill residues for particleboard furnish will be tight.

#### FUTURE EXPECTATIONS FOR BONDED WOOD PRODUCTS

Ву

Thomas H. Ellis, Economist Forest Products Laboratory Madison, Wisconsin

The title of my paper may suggest to you that I have access to an oracle somewhere who knows what the future will be. To correct this notion, I will start hedging immediately, somewhat as the Census Bureau did in reporting its recent population projections.

The Bureau distinguished between projections, predictions, and forecasts. Projections are merely estimates of what might happen if certain assumptions held true. Predictions are the kinds of statements one would make if he really knew what was going to happen. And forecasts are sort of inbetween projections and predictions, being estimates with statements as to the likelihood of their being correct. I think I have the distinction figured out now. Today, I'm giving you some projections. If they turn out right, in a few years I'll call them predictions.

# Outline |

First I want to mention briefly some broad trends in lumber and wood products, then present some demand projections published last month by the Forest Service, and finally to discuss some of the major uncertainties regarding those projections.

For presentation at the 1975 Symposium on Adhesives for Products from Wood, Wisconsin Center, Madison, WI. September 24-26, 1975.

#### Broad Trends

I'm sure you all know what a broad trend is. That's what happened to your wife between her 20th and 40th birthdays. You're probably equally aware of the weighty shifts in forest products over the past 20 years, but I want to list a few of them anyway.

Probably the most important trend in U.S. wood products markets has been the displacement of solid lumber by plywood--not because plywood was cheaper on a volume basis but because its in-place costs for many applications were lower than for lumber.

Then we have had displacement of plywood by particleboard which tended to be less costly on a volume basis and also had characteristics which gave it a competitive edge in some furniture applications and in uses where large panels were desirable--as in mobile home flooring systems.

More recently, medium density fiberboard has appeared as a competitor to particleboard, with advantages of even greater flexibility in using various kinds of mill residues, as well as superior physical characteristics in furniture uses.

We have also seen the entry of thin particleboard, composite particleboard veneer panels, structural particleboards, panels with alined fibers, and proposals for reconstituted lumber-type products such as Press-lam and Com-Ply.

Imagine the frustration of the Census Bureau in classifying forest products for the next Census of Manufacturers.

Another major factor in forest products markets was the large changes in housing construction mix, with an increasingly large proportion in multi-family units and mobile homes. Single-family units grew increasingly large and expensive, but lumber use per square foot declined.

The result of all of these shifts in use was that total U.S. lumber consumption stayed virtually constant between 1950 and 1970, despite a doubling of real GNP and a 1/3 increase in population. But consumption of reconstituted wood products increased dramatically.

## Recent Forest Service Projections

The most recent Forest Service projection of forest products demand and supply are those published last month in the Draft Assessment Report prepared as required by the Forest and Rangeland Renewable Resources Act of 1974. These were slightly modified versions of those presented in the 1973 report, "The Outlook for Timber in the United States."

The new projections indicated substantial increases in demand for softwood and hardwood lumber, mainly due to housing construction and pallet production.

A 56 percent increase in plywood demand was projected for 1980, as compared to 1970, with half of this increase attributed to housing.

1980 building board consumption, including insulation board, hardboard and particleboard was expected to be 62 percent above 1970 with large increases in usage for manufacturing and new housing.

Demand for particleboard was projected to be 4.1 billion square feet in 1980 and 5.7 billion in 1990.

The Forest Service estimates for particleboard appear quite conservative as compared to ones made recently by Leonard Guss. Dr. Guss projected 9.5 billion square feet for 1983. The major reason for the difference appears to be that the Forest Service assumed that substitution of panel products for lumber in housing was nearly at an end. Also, I believe Dr. Guss assumed a much more rapid implementation of structural particleboard, at the expense of plywood, than did the Forest Service.

## Assumptions

The demand projections I've discussed were based on assumptions regarding general technological, economic and population trends. For example, materials use in housing was estimated by first analyzing the effect of population changes on volume and type of housing, then projecting changes in floor space per unit and in wood products use per square foot of floor space.

The population analysis, made by Tom Marcin of the North Central Forest Experiment Station, was published earlier this year. Considering current age-class distribution it appears that a bulge in the population will move through the 30-44 year old age classes in the late 1970's and in the 1980's--probably causing a rise in demand for detached single-family homes. Marcin expected a large demand for mobile homes and multi-family homes into the early 1980's due to large numbers of households headed by people below age 30. By 1990, a large decrease in demand could occur due to recent low birth rates.

In 1974, new for sale 1-family detached homes surveyed by NAFB, averaged almost 1,700 square feet of floor space. The Forest Service projections were based on 1,615 square feet for 1980 and 1,680 for 1990.

For products used in nonresidential construction, shipping and manufacturing, projections were based largely on assumed relationships between GNP and economic activity within each sector, and on trends in wood products use relative to total spending in each sector. Annual rates of increase in GNP (in constant dollars) were assumed to average 4 percent through the 1970's and 3.5 percent in the 1980's.

## Major Uncertainties

The projections just described here give a fairly encouraging outlook for forest products for the next 10-15 years. There could be some nasty flies in the ointment however, mainly in regard to uncertainties about the economy in general and upon housing in particular. In recent years, prices of new housing have risen much faster than has per capita disposable income. High mortgage rates coupled with soaring construction costs may mean a return to smaller housing units. Some people have predicted increased demand for multi-family housing as a means of reducing land costs and heating bills.

Resource problems undoubtedly will continue to affect forest products industries. Two trends here seem particularly important. One is the continuing shift from western timber to southern supplies.

Southern pine inventories increased substantially in the 1950's and 1960's while Pacific coast inventories declined. National forest inventories on the Pacific coast are still huge, but public controversy over National Forest management is likely to limit chances for increased timber harvesting there in the next 10 years or so.

The second major trend in wood resources has been the tremendous increase in consumption of sawmill and veneer mill residues by pulp and board manufacturers. Some crude estimates I made for 1973 indicate that about 59 million tons of wood residues were produced at mills in 1973, but only 10 million tons went unused. Probably 60 percent was used for pulp and particleboard and another 10-15 percent was used for fuel or miscellaneous products. Improvements in sawmilling and veneer manufacturing could result in reduced total production of residues by 1990, while fuelwood values increase. Competition for mill residues is bound to mean higher prices for sawdust, planer shavings, and chips.

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The alternative seems likely to be greater use of forest residue-type materials or of hardwoods (SLIDE 12). Increases in hardwood inventories have more than offset decreases in softwood sawtimber over the past 20 years.

## Conclusions

In concluding, let me confess that I am unable to resolve the major uncertainties posed by energy problems and general economic conditions. If you are pessimistic about the general economic outlook, then the Forest Service projections of panel products demand will be too high for you. If you are an economic optimist, the Forest Service projections of total panel products consumption may look too low.

In any case, if there are to be large increases in particleboard, MDF, or hardboard production, most of the increase in raw material will have to come from roundwood or from the type of material formerly left as logging residue. And, these industries will have to develop more efficient logging methods and more efficient adhesive systems if they want to compete with plywood.

#### APPENDIX

## MEDIUM PROJECTIONS OF DEMAND FOR FOREST PRODUCTS

#### from

ASSESSMENT (Draft) -- The Nations Renewable Resources -- An Assessment, 1975, as required by the Forest and Range and Renewable Resources Planning Act of 1974. USDA, Forest Service, August 1975.

## MEDIUM PROJECTIONS OF DEMAND

#### Lumber

		Softwoods Board Feet)
1970 Consumpti	n 7.3	32.1
1973 Consumpti	n 7.1	38.7
1980 Projectio	9.4	42.8
1990 Projectio	10.8	46.0

## MEDIUM PROJECTIONS OF DEMAND

## Lumber

	1970 Consumption	Increase by 1980
New Housing	(Billion B.F.) 12.3	% 42
Residential Upkeep & Repair	4.7	17
New Nonresidential Construction	3.7	30
Manufacturing	4.7	26
Shipping	5.7	35
All Other	8.5	28
Total	39.6	33

# MEDIUM PROJECTIONS OF DEMAND

# Plywood

	1970 Consumption	Increase by 1980
	Billion Sq. Ft. (3/8 <sup>th</sup> Basis)	%
New Housing	6.3	66
Residential Upkeep and Repair	2.5	33
New Nonresidential Construction	1.7	63
Manufacturing	1.7	45
All Other	5.6	<u>56</u>
Total	17.8	56

# MEDIUM PROJECTIONS OF DEMAND

# Building Board

	1970 Consumption	Increase by 1980
	Billion Sq. Ft. (3/8" Basis)	8
New Housing	2.8	53
Residential Upkeep and Improvements	1.4	34
New Nonresidential Construction	1.0	36
Manufacturing	1.8	94
All Other Uses	2.7	74
Total	9.7	62

# MEDIUM PROJECTIONS OF DEMAND

# Panel Products

		Increase	Increase
	1973 Consumption	by 1980	by 1990
Softwood Plywood	17.8 Billion Sq. Ft.,	%	%
,	3/8"	24	43
Hardwood Plywood	4.9 Billion Sq. Ft., 3/8"	26	46
Particleboard	3.5 Billion Sq. Ft.,		
railicleboald	3/4"	19	65
Hardboard	2.4 Million Tons	11	63
Insulation Board	1.7 Million Tons	-12	<del>-</del> 3

# NEW HOUSING PRODUCTION - AVERAGE ANNUAL TOTAL

## 1000's of Units

	Actual	1973	1975
Period	Starts	Projections	Projections
1950-59	1520		
1960-69	1650		
1970-79		2,500	2,460
1980-89		2,620	2,560
1990-99		2,560	2,360

# NEW HOUSING PRODUCTION - AVERAGE ANNUAL

# 1000's of Units

	Actual		1973 Projections			
Period	I & 2 Family	Multi- Family	Mobiles	I & 2 Family	Multi- Family	Mobiles
1950-59	1320	140	60			
1960-69	1440	980	460			
1970-79				1160	810	530
1980-89				1410	690	520
1990-99				1460	610	490

#### HAL ROTH

Hal is a Special Accounts Manager for E.I. DuPont de Nemours and Co., Inc. He received his academic training in polymer chemistry at the University of Akron. Between 1947 and 1953 he carried out research on polymers with the Goodyear Tire and Rubber Co. He has been with DuPont since 1953, where he was first involved in research and development of urethane foams and cast polymer systems. For the past 8 years he has been involved in selling and marketing research activities.

#### FUTURE OF THE ADHESIVES INDUSTRY

This report is a brief summary of an Interindustry Delphi study on the Future of the Adhesives Industry in the United States. The Delphi Study consisted of three rounds of questioning with some level of participation recorded by 382 individuals representing 131 separate organizations. Assessment was made of the general changes that might take place in the adhesives industry as well as the use of adhesives in residential construction, in furniture assembly, in prefabricated metal products, in transportation equipment, in textile products and apparel, in footwear, and in packaging and packaging materials. This report emphasizes the general changes in the industry and adhesive applications on wood and wood products. The discussion of general changes will include the effects of government regulations and material shortages, hindrance to or impetus for new developments, and changes in adhesive characteristics. Factors affecting the acceptance of adhesive bonding in residential construction will be discussed along with progress toward adoption of adhesive bonding, adhesives in specific applications, and types of adhesives that may be used in the future.

#### JOHN TOOGOOD

John is a Market Manager with the Chemicals and Plastics Division of Union Carbide Corporation in New York, with responsibility for adhesive and sealant intermediates. He was born in Cardiff, Wales and educated at the University of London, where he received his Ph. D. in Organic Chemistry. After working for Imperial Chemical Industries in Yorkshire, England, he emigrated to Canada, working with Canadian Industries Limited on polyester fiber production. In 1955, John came to the United States and joined Union Carbide in product development. He later was in technical service with the Silicones Division and had a marketing assignment on the West Coast, before he took his present position.

## FUTURE OF PETROCHEMICALS FOR ADHESIVES

The U.S.-based adhesives industry is a large user of a complex of chemical raw materials ranging from simple molecules like styrene and vinyl acetate through sophisticated chemicals and polymers. These amount to about 50% of materials used, the other 50% being natural products.

The majority of the synthetic raw materials are petroleum-based. The petrochemical industry which supplies all the hydrocarbon-based raw materials depends almost entirely upon naphtha, a distillate of crude oil.

The wood products industry has a high dependency upon phenol and vinyl acetate, which in turn means a high dependency upon the availability of propylene and ethylene. Over the next 5 years, no shortage of propylene is anticipated because of the projected reduced demand for gasoline. Additions to capacity already under construction are also expected to fully meet the projected increased demands for ethylene.

Since the Arab oil crisis, the petrochemical industry has been forced into a reevaluation of its dependency upon crude oil. One of the primary objectives for new technology is to free the industry from its dependence on ethylene for most of its oxygenated materials. This in turn, should free them from their dependency on petroleum.

A more complete presentation is given in Twogood's "The Future of Petrochemicals for Adhesives," published in Adhesives Age, January 1976.









